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# Data Summary Report for Fission Product Release Test VI-1

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Prepared by M. F. Osborne, J. L. Collins, R. A. Lorenz,  
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**Oak Ridge National Laboratory**

**Prepared for  
U.S. Nuclear Regulatory  
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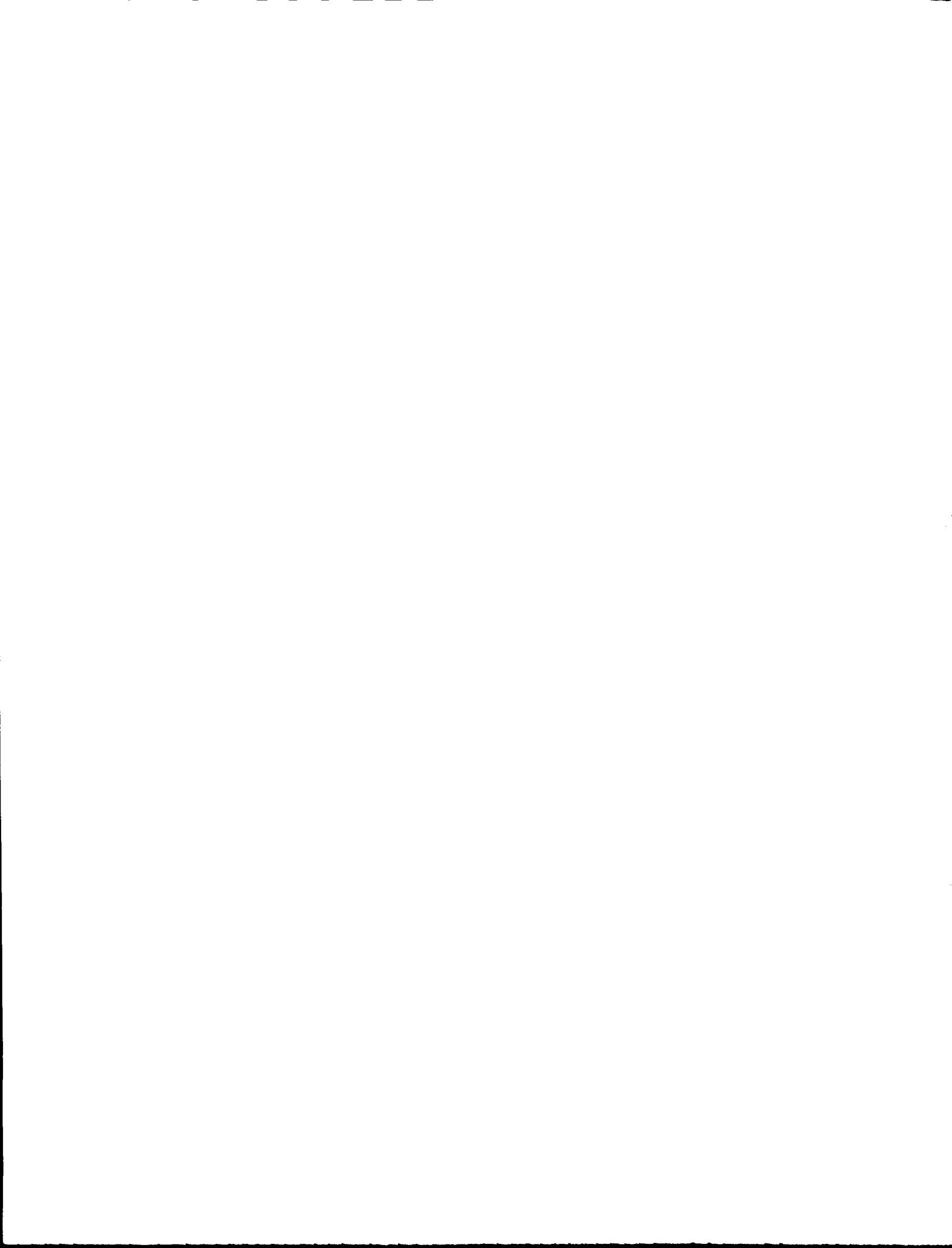
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## ABSTRACT

The first in a series of high-temperature fission product release tests in a new vertical test apparatus was conducted in flowing steam. The test specimen was a 15.2-cm-long section of a fuel rod from the Oconee 1 PWR; it had been irradiated to a burnup of ~42 MWd/kg. Using an induction furnace, it was heated under simulated LWR accident conditions — 20 min at 2000 K and 20 min at 2300 K — in a hot cell-mounted test apparatus.

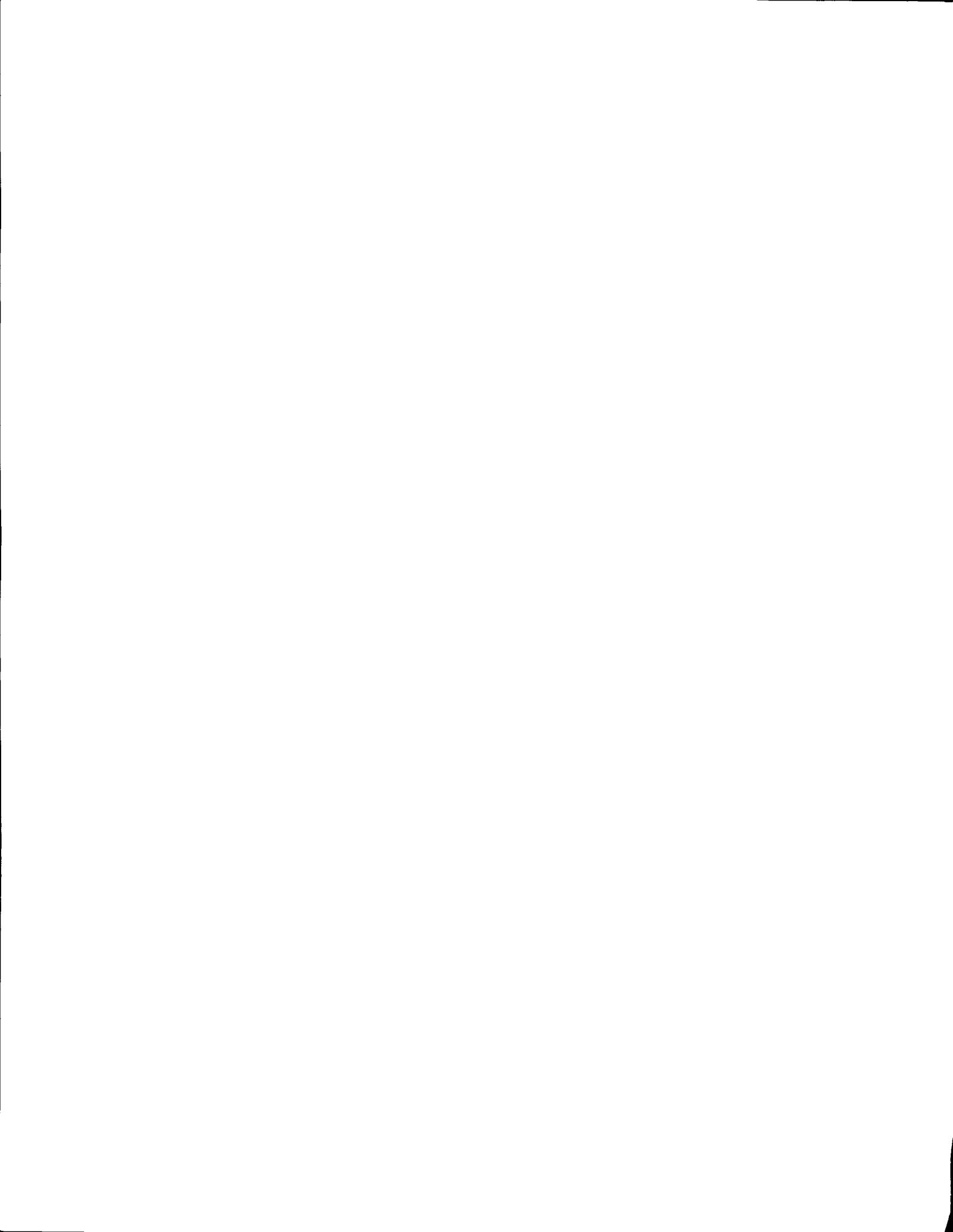
Posttest inspection showed severe oxidation but only minimal fragmentation of the fuel specimen; cladding melting was apparent only near the top end. Based on fission product inventories measured in the fuel and/or calculated by ORIGEN, analyses of test components showed total releases from the fuel of 47% for  $^{85}\text{Kr}$ , 33% for  $^{125}\text{Sb}$ , 37% for  $^{129}\text{I}$ , 84% for  $^{110\text{m}}\text{Ag}$ , and 63% for  $^{137}\text{Cs}$ . Large fractions (36% and 30%, respectively) of the released  $^{110\text{m}}\text{Ag}$  and  $^{125}\text{Sb}$  were retained in the furnace above the fuel. Pretest and posttest analysis of the fuel specimen indicated a  $^{134}\text{Cs}$  release of 65%, which is very good agreement with the  $^{137}\text{Cs}$  value.

The first phase (2000 K) of this test was conducted at the same conditions as test HI-2. However, the releases of Cs, I, and Kr were much less than those in HI-2. Variations in fuel characteristics (H. B. Robinson in test HI-2 vs Oconee in test VI-1) are believed to be major factors in this difference, and the large axial crack that occurred in HI-2 probably was a significant factor.



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## FOREWORD

This document describes the seventh in a series of fission product release tests of commercial LWR fuel. Other reports describing work under this fission product release program are:

1. M. F. Osborne, R. A. Lorenz, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-1, NUREG/CR-2928 (ORNL/TM-8500), December 1982.
2. M. F. Osborne, R. A. Lorenz, J. R. Travis, C. S. Webster, and K. S. Norwood, Data Summary Report for Fission Product Release Test HI-2, NUREG/CR-3171 (ORNL/TM-8667), February 1984.
3. M. F. Osborne, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-3, NUREG/CR-3335 (ORNL/TM-8793), April 1984.
4. M. F. Osborne, J. L. Collins, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-4, NUREG/CR-3600 (ORNL/TM-9001), June 1984.
5. M. F. Osborne, J. L. Collins, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-5, NUREG/CR-4037 (ORNL/TM-9437), May 1985.
6. M. F. Osborne, J. L. Collins, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-6, NUREG/CR-4043 (ORNL/TM-9943), September 1985.
7. M. F. Osborne, J. L. Collins, R. A. Lorenz, J. R. Travis, and C. S. Webster, Design, Construction, and Testing of a 2000°C Furnace and Fission Product Collection System, NUREG/CR-3715 (ORNL/TM-9135), September 1984.
8. J. L. Collins, M. F. Osborne, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Observed Behavior of Cesium, Iodine, and Tellurium in the ORNL Fission Product Release Program, NUREG/CR-3930 (ORNL/TM-9316), February 1985.
9. K. S. Norwood, An Assessment of Thermal Gradient Tube Results from the HI Series of Fission Product Release Tests, NUREG/CR-4105 (ORNL/TM-9506), March 1985.
10. M. F. Osborne, J. L. Collins, P. A. Haas, R. A. Lorenz, J. R. Travis, and C. S. Webster, Design and Final Safety Analysis Report for Vertical Furnace Fission Product Release Apparatus in Hot Cell B, Building 4501, NUREG/CR-4332 (ORNL/TM-9720), March 1986.

11. M. F. Osborne, J. L. Collins, and R. A. Lorenz, Highlights Report for Fission Product Release Tests of Simulated LWR Fuel, ORNL/NRC/LTR-85/1, February 1985.
12. M. F. Osborne, J. L. Collins, R. A. Lorenz, and T. Yamashita, Highlights Report for Fission Product Release Test VI-1, ORNL/NRC/LTR-86/7, March 1986.
13. M. F. Osborne, J. L. Collins, R. A. Lorenz, and T. Yamashita, Highlights Report for Fission Product Release Test VI-2, ORNL/NRC/LTR-86/18, January 1987.
14. M. F. Osborne, J. L. Collins, R. A. Lorenz, J. R. Travis, C. S. Webster, S. R. Daish, H. K. Lee, T. Nakamura, and Y.-C. Tong, Highlights Report for Fission Product Release Test VI-3, draft letter report to Severe Fuel Damage Program partners, August 1987.
15. S. K. Wisbey, Preliminary Studies of the Morphology of Thermal Gradient Tube Deposits for Fission Product Release Experiments, NUREG/CR-4778 (ORNL/TM-10273), March 1988.
16. T. Yamashita, Steam Oxidation of Zircaloy Cladding in the ORNL Fission Product Release Tests, NUREG/CR-4777 (ORNL/TM-10272), March 1988.

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The authors gratefully acknowledge the significant contributions of several colleagues in conducting this work: C. W. Alexander for assistance with ORIGEN2 calculations of fission product inventories; D. A. Costanzo and co-workers of the Analytical Chemistry Division for the  $^{129}\text{I}$  and U determinations and spark-source mass spectrometry; H. K. Lee of Korea — Advanced Energy Research Institute, T. Nakamura of Japan-JAERI, and Y.-C. Tong of Taiwan — Institute of Nuclear Energy Research, for assistance with analysis and interpretation of data; C. A. Culpepper of the Isotopes Division and S. J. Wisbey of UKAEA-Harwell for SEM/EDX examination and analysis; E. C. Beahm and T. B. Lindemer for technical consultation; B. C. Drake for preparation of the manuscript; and M. L. Conte for editing.



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DATA SUMMARY REPORT FOR FISSION PRODUCT RELEASE FROM TEST VI-1

M. F. Osborne	J. R. Travis
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R. A. Lorenz	T. Yamashita

1. EXECUTIVE SUMMARY

The objective of this report is to document the observations and results of this fission product release test (VI-1). Complete interpretation and correlation of these results with related experiments and with theoretical behavior will be included in a subsequent report, which will consider the results of several tests over a range of test conditions. Similar data summary reports for previous tests in this series are listed in the Foreword.

The fuel specimen used in this test was cut from a standard fuel rod irradiated in the Oconee 1 Pressurized-Water Reactor (PWR) from February 1975 to November 1979. The fabrication and irradiation history of this specimen is detailed, and the fission product inventories, as measured in the fuel and calculated by ORIGEN2, are listed in Sect. 3. The test results and some preliminary interpretations are presented in Sect. 4. The most important results are:

1. This was the first test in a new, vertically-oriented induction furnace with three sequentially-operated fission product collection trains. Other new test equipment included a heated CuO bed, a condenser, and a modified pH meter for the conversion and continuous measurement of hydrogen generated by the reaction of steam with the Zircaloy cladding on the fuel specimen. With the exception of overloading of some of the on-line radiation detectors (saturation caused by higher-than-expected levels of radiation), all of the experimental apparatus performed quite well.
2. The test conditions were more severe than in any of our previous tests of high-burnup commercial fuel. The three test phases, one for each collection train, were (1) heatup and 20 min at 2020 K; (2) further heatup and 5 min at 2300 K; and (3) an additional 15 min at 2300 K, followed by cooldown, all in a steam-helium mixture flowing at ~1.5 L/min steam and ~0.4 L/min helium at STP.
3. Posttest examination showed that the Zircaloy cladding was completely oxidized, contained numerous minor fractures, and had experienced limited melting and/or fuel-cladding interaction. The 15-cm-long specimen, however, did not collapse but remained in its original vertical orientation. The UO<sub>2</sub> fuel exhibited a significant increase in porosity, apparently the result of extensive bubble formation, growth, and interlinkage as the volatile fission products escaped from the matrix. Measurements of the increased porosity, however, were not made.

4. The oxidation behavior of the Zircaloy cladding, as indicated by hydrogen generation, was in good agreement with the Zircaloy oxidation model, and with generally accepted literature values for the oxidation rates as a function of temperature.
5. Techniques for determining the inventories of fission products in the irradiated fuel were significantly improved, thereby allowing more accurate release measurements. Using the concentrations of the long-lived gamma emitters ( $^{106}\text{Ru}$ ,  $^{120}\text{Sb}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{144}\text{Ce}$ , and  $^{145}\text{Eu}$ ) that were measured by gamma spectrometry, and neodymium and uranium isotopic data, which were obtained by mass spectroscopy, ORIGEN2 calculations were used to produce improved values for all important fission product nuclides and elements.
6. The values for total release from the fuel specimen for  $^{85}\text{Kr}$ ,  $^{110\text{m}}\text{Ag}$ ,  $^{125}\text{Sb}$ ,  $^{129}\text{I}$ , and  $^{137}\text{Cs}$  were 47, 84, 33, 37, and 73% respectively. About one-third of the released  $^{110\text{m}}\text{Ag}$  and  $^{125}\text{Sb}$ , however, were deposited on the  $\text{ZrO}_2$  ceramics at the outlet end of the furnace, compared to almost complete transport of the released Kr, I, and Cs from the furnace to the collection system.
7. Determination of the fission product distributions in the thermal gradient tubes showed a large cesium peak at  $\sim 500^\circ\text{C}$  and a small Sb peak at the entrance ( $\sim 850^\circ\text{C}$ ) of tube A; a large cesium peak at  $\sim 750^\circ\text{C}$  in tube C; and Sb peaks at the entrances, followed by continuous low levels of Sb in both tubes B and C. After leaching to remove the soluble deposits, cesium, antimony, and silver were measured throughout all three tubes, apparently alloyed or insolubly combined with the platinum deposition tubes, in approximately logarithmically decreasing concentrations.
8. Small samples of the deposits on the thermal gradient tubes (TGTs) were examined and analyzed by scanning electron microscope (SEM) with energy-dispersive x-ray analysis (EDX). Cesium was the only fission product identified, but structural and impurity elements were found.
9. The masses of deposits on the TGTs and filters were determined by direct weighing. A total of 1.17 g of material was found, with almost half (0.52 g) being deposited during the last (2300 K) phase of the test.

## 2. INTRODUCTION

This report summarizes data from the first test in a new vertical test apparatus. This series is designed to investigate fission product release from light-water reactor (LWR) fuel in steam and/or hydrogen in the temperature range of 2000 to 2700 K. Earlier tests, conducted under similar conditions at temperatures of 773 to 1873 K (500 to 1600°C), have been reported by Lorenz et al.<sup>1-4</sup> The purpose of this work, which is sponsored by the U.S. Nuclear Regulatory Commission (NRC), is to obtain the experimental data needed to reliably assess the consequences of a

variety of heatup accidents in LWRs.\* The primary objectives of this program are:

1. to determine fission product release from commercial LWR fuel at temperatures up to and including fuel melting (~2700 K) in steam and/or hydrogen atmospheres;
2. to identify, to the extent possible, the chemical and physical forms of the released fission products;
3. to compare the observed fission product behavior with the physical and chemical changes in the fuel specimens; and
4. to compare the results with data from related programs and to develop a consistent source term model applicable to any LWR fuel subjected to a spectrum of accident conditions.

Tests of high-burnup LWR fuel are emphasized in this program. The applicability of simulated fuel (unirradiated  $UO_2$  containing fission product tracers) will be considered where appropriate. All tests will be conducted in a flowing mixture of steam and helium, or hydrogen and helium, at atmospheric pressure; steam concentrations will be varied to simulate different accident sequences or core locations.

This vertical test apparatus<sup>5</sup> expands test capabilities over the previous horizontal equipment<sup>6</sup> in several ways: (1) the furnace and TGTs are vertical, more nearly simulating the orientation of the fuel rods in an LWR; (2) three collection trains, each including a TGT and filter package, may be used in sequence during the test to provide better time and temperature resolution; (3) temperature measurement and control have been improved, limiting axial thermal gradients over the fuel specimen to <50 K; and (4) gas flow rate and distribution have been modified to provide more uniform atmospheric (oxidizing and reducing) conditions over the length of the specimen. The procedures and techniques used in posttest examination and analysis were very similar to those used for the six HI tests.<sup>7-12</sup> The analytical techniques employed are listed in Table 1, and the general procedure for preparing and conducting a fission product release test is shown in the Appendix.

This report provides a brief description of test VI-1 and a tabulation of all the results obtained. Thorough data evaluation and correlation with other test results will be included in subsequent reviews and reports covering this series of fission product release tests at temperatures up to 2700 K.

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\*M. F. Osborne, R. A. Lorenz and R. P. Wichner, "Program Plan for Fission Product Release From LWR Fuel in Steam," memorandum to USNRC, April 1982.

Table 1. Analytical techniques for fission product analysis

Technique	Time	Location	Elements
Gamma spectrometry	Pretest, posttest	Fuel specimen	Long-lived, gamma-emitting fission products — Ru, Ag, Sb, Cs, Ce, Eu
	On-line	Thermal gradient tube, charcoal traps, filters	Cs, Kr
	Posttest	Furnace components, thermal gradient tube, filters	Ru, Ag, Sb, Cs, Ce, Eu
Activation analysis	Posttest	Charcoal, solution from furnace, thermal gradient tube, filters	I, Br, (Te, Mo, Ba) <sup>a</sup>
Chemical analysis	Posttest	Furnace, filters	U, Pu
Spark-source mass spectrometry	Posttest	Samples from furnace, thermal gradient tube, filters	All elements with atomic numbers >10
Scanning electron microscope, with energy dispersive x-ray system	Posttest	TGT, aerosol sampler	All elements with atomic numbers >10

<sup>a</sup>Iodine and bromine have been analyzed by neutron activation throughout HI and VI test series; neutron activation analysis techniques for Te, Mo, and Ba are in various stages of development.

## 3. DESCRIPTION OF TEST VI-1

The series of tests planned for this vertical apparatus includes temperature (2000 to 2700 K) as the dominant variable; steam flow rate (0.3 to 1.5 L/min) and time (1 to 60 min), both of which affect the extent of oxidation, are secondary variables. A tentative test matrix, which defines the conditions for a series of eight tests, is shown in the Appendix. The objective of this particular test was to obtain release data at 2000 and 2300 K for 20 min each in steam flowing at ~1.5 L/min. The first phase of this test (2000 K) provided conditions comparable to tests HI-2 and HI-5.

## 3.1 FUEL SPECIMEN DATA

The test specimen was a 15.2-cm-long section of rod 08747 from a 15 x 15 fuel assembly (serial No. 1D13), which operated in the Oconee Unit 1 reactor from Feb. 11, 1975, to Nov. 22, 1979.<sup>13</sup> Details of the irradiation and the characteristics of this particular specimen are listed in Table 2. Fission product inventories for the specimen as calculated by Alexander\* with ORIGEN2<sup>14</sup> are shown in Tables 3 and 4. A small sample of fuel, cut from the rod adjacent to the test specimen, was dissolved and analyzed by mass spectrometry. These data, shown in Table 5, were used to determine the burnup of the fuel, a critical input for the ORIGEN2 calculation.

Table 2. Data for fuel specimen used in test VI-1

Fuel rod identification	Rod 08747 Assembly 1D13 Oconee Unit 1 (PWR)
Irradiation data	
Period	Feb. 11, 1975 to Nov. 22, 1979 (four cycles)
Fuel specimen burnup <sup>a</sup>	~40 MWd/kg U
Specimen data	
Length	15.2 cm
Fuel loading	109.18 g UO <sub>2</sub> (96.24 g U)
Total weight	140.36 g
Weight of Zircaloy	31.18 g
Gas release during irradiation	0.52% (from whole rod) 0.7% (from specimen)

<sup>a</sup>The reported burnup values varied between 38 and 42 MWd/kg U depending on measurement method. (Values taken from Ref. 14: G. M. Bain and W. A. McInteer, Data Package for High Burnup Fuel Samples, LRC 4063, Lynchburg Research Center, Babcock and Wilcox Co., January 1983.

\*C. W. Alexander, ORNL, Personal Communication, June 1984.

Table 3. Inventories of principal fission product elements  
in test VI-1 fuel<sup>a</sup>

Elements	Amounts in fuel (g/MTU)	Amounts in <sup>b</sup> test specimen (g) $\times 10^3$
Se	66.5	6.40
Br	25.3	2.44
Kr	424.0	40.81
Rb	399.0	38.40
Sr	977.4	94.06
Y	525.0	50.53
Zr	4165.0	400.84
Mo	4007.0	385.63
Tc	913.8	87.94
Ru	2771.0	266.68
Rh	517.6	49.81
Pd	1832.0	176.31
Ag	98.2	9.45
Cd	153.0	14.72
In	2.5	0.24
Sn	113.6	10.93
Sb	32.0	3.08
Te	589.1	56.69
I	288.1	27.73
Xe	6425.0	618.34
Cs	3160.0	304.12
Ba	1839.0	176.99
La	1453.0	139.84
Ce	2883.0	277.46
Eu	198.4	19.09
Total fission products	41140.0	3959.31
U	947900	91226
Pu	9992	962
Total actinides	958900	92285

<sup>a</sup>Inventories for this Oconee fuel were calculated by ORIGEN2, assuming a burnup of 40,000 MWd/MTU, and were adjusted based on the results of gamma ray spectrometric analysis of the fuel before testing, corrected for decay to July 15, 1981.

<sup>b</sup>The test VI-1 specimen contained 96.24 g U =  $9.624 \times 10^{-5}$  MTU before testing.

Table 4. Inventory of long-lived radionuclides in test VI-1 fuel<sup>a</sup>

Radionuclides	Amounts in fuel		Amounts in test specimen <sup>b</sup>	
	(g/MTU)	(Ci/MTU) <sup>c</sup>	(mg)	(Ci) <sup>c</sup>
<sup>85</sup> Kr	23.05	9048	2.22	0.871
<sup>90</sup> Sr	573.50	78250	55.19	7.531
<sup>106</sup> Ru	47.57	159200	4.58	15.321
<sup>110m</sup> Ag	0.19	907	0.02	0.087
<sup>125</sup> Sb	9.39	9697	0.90	0.933
<sup>129</sup> I	218.20	0	21.00	0.000
<sup>134</sup> Cs	81.56	105600	7.85	10.163
<sup>137</sup> Cs	1355.00	117900	130.41	11.347
<sup>144</sup> Ce	55.67	177700	5.36	17.102
<sup>154</sup> Eu	46.73	12620	4.50	1.215
Total fission products	41140.00	1290000	3959.31	124.150
<u>Actinides</u>				
<sup>234</sup> U	152.4	0.9525	15	0.00009
<sup>235</sup> U	5502	0.0119	530	0.00000
<sup>236</sup> U	4344	0.2812	418	0.00003
<sup>238</sup> U	937900	0.3155	90263	0.00003
Total U	947900	4.969	91226	0.00048
<sup>238</sup> Pu	249.5	4273	24	0.41123
<sup>239</sup> Pu	5114	318	492	0.03060
<sup>240</sup> Pu	2615	596.1	252	0.05737
<sup>241</sup> Pu	1336	137700	129	13.25225
<sup>242</sup> Pu	676.9	2.585	65	0.00025
Total Pu	9992	142900	962	13.7570
Total actinides	958900	153400	92285	14.76322

<sup>a</sup>Inventories for this Oconee fuel were calculated by ORIGEN2, assuming a burnup of 40,000 MWd/MTU, and were adjusted based on the results of gamma ray spectrometric analysis of the fuel before testing.

<sup>b</sup>The test VI-1 specimen contained 96.24 g U = 9.624E-05 MTU before testing.

<sup>c</sup>Corrected for decay to July 15, 1981.

Table 5. Mass spectrometric data used to determine burnup  
in Oconee fuel rod 08747 (test VI-1)

Nuclide	Concentration		Mass of nuclide (mg/g fuel)
	(atm %)	(wt %)	
<u>Neodymium</u>			
<sup>142</sup> Nd	0.646	0.633	0.0276
<sup>143</sup> Nd	18.105	17.862	0.7802
<sup>144</sup> Nd	34.141	33.919	1.4816
<sup>145</sup> Nd	16.133	16.139	0.7050
<sup>146</sup> Nd	17.538	17.666	0.7716
<sup>148</sup> Nd	9.045	9.236	0.4034
<sup>150</sup> Nd	4.392	4.545	0.1985
Total	100	100.000	4.368
mg Nd/g fuel	4.368		
<u>Uranium</u>			
<sup>234</sup> U	0.0157	0.015	0.1309
<sup>235</sup> U	0.608	0.600	5.0897
<sup>236</sup> U	0.472	0.468	3.9680
<sup>238</sup> U	98.904	98.916	838.5115
Total	99.9997	100.000	847.700
mg U/g fuel	847.7		
<u>Plutonium</u>			
<sup>238</sup> Pu	2.766	2.746	0.2571
<sup>239</sup> Pu	50.74	50.588	4.7361
<sup>240</sup> Pu	26.252	26.283	2.4606
<sup>241</sup> Pu	12.366	12.432	1.1639
<sup>242</sup> Pu	7.875	7.950	0.7443
Total	99.999	100.000	9.362
mg Pu/g fuel	9.362		

An axial scan of the total gamma radioactivity along the fuel rod is shown in Fig. 1. Reductions in burnup resulting from the flux depression caused by the grid spacers are apparent. This gamma scan is dominated by  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , but also includes the emissions from such long-lived, gamma-emitting nuclides as  $^{106}\text{Ru}$ ,  $^{110\text{m}}\text{Ag}$ ,  $^{125}\text{Sb}$ ,  $^{144}\text{Ce}$ , and  $^{154}\text{Eu}$ . The total gas release (Kr and Xe) from the entire fuel rod was reported to be 0.52%.<sup>14</sup> Since very little gas could have been released from the lower-burnup, lower-temperature regions near the ends of the rod, we assume that all gas released came from the high-burnup region between 20 and 120 in. The test specimen was cut from the high-burnup region of the rod, as indicated in Fig. 1. We calculated that ~0.7% of the krypton and xenon was released from the test specimen during irradiation, and consequently, was not available for this test.

In addition to the test VI-1 fuel specimen, three short samples (1 to 2 cm long) were cut from section 10 for metallographic examination and chemical analysis, and a 19-cm-long section was retained for possible future use.

Tapered end caps of Zircaloy-2 were pressed onto the ends of the test specimen, not as gas seals, but to prevent loss of the fractured  $\text{UO}_2$  fuel during subsequent handling. In addition, a second end cap with a pin to facilitate vertical mounting was pressed onto the bottom end of the specimen. A small hole, 1.6 mm in diameter, was drilled through the cladding at midlength to serve as a standard leak for gas release during the heatup phase of the test. These details are shown in Fig. 2.

### 3.2 EXPERIMENTAL APPARATUS

The fuel specimen was heated in an induction furnace, as illustrated in Fig. 3. This furnace was developed from the design used in the previous fission product release tests.<sup>6</sup> The graphite susceptor was protected from extensive steam oxidation by a blanket of helium gas that leaked through joints in the  $\text{ZrO}_2$  ceramics into the flowing helium-steam mixture (see Fig. 3). The furnace was mounted inside a stainless steel containment box in a hot cell, as shown in Fig. 4. The fission product collection system included platinum TGTs, graduated fiberglass filters, heated charcoal (for iodine adsorption), and cooled charcoal (for inert fission gas adsorption). The steam was collected in a condenser and a dryer, as indicated in Fig. 5, prior to reaching the cooled charcoal. Instrumentation included a W-5% Re vs W-26% Re thermocouple installed in a ceramic thermowell in the inlet end of the furnace (Fig. 3); a manually operated optical pyrometer of the disappearing-filament type and two automatic two-color pyrometers, all located outside the hot cell as indicated in Fig. 4; one Ge(Li) and two NaI(Tl) radiation detectors connected to a multichannel analyzer; infrared analyzers for CO and  $\text{CO}_2$  measurement and a CuO bed with a condenser/conductivity cell for  $\text{H}_2$  measurement in the collection system, as shown in Fig. 5; and conventional electrical and gas flow instruments. A data acquisition system (Fig. 6) was used to record test data at 1-min intervals, and several individual chart recorders maintained continuous records of temperatures and flow rates. Differences in apparatus materials and conditions for

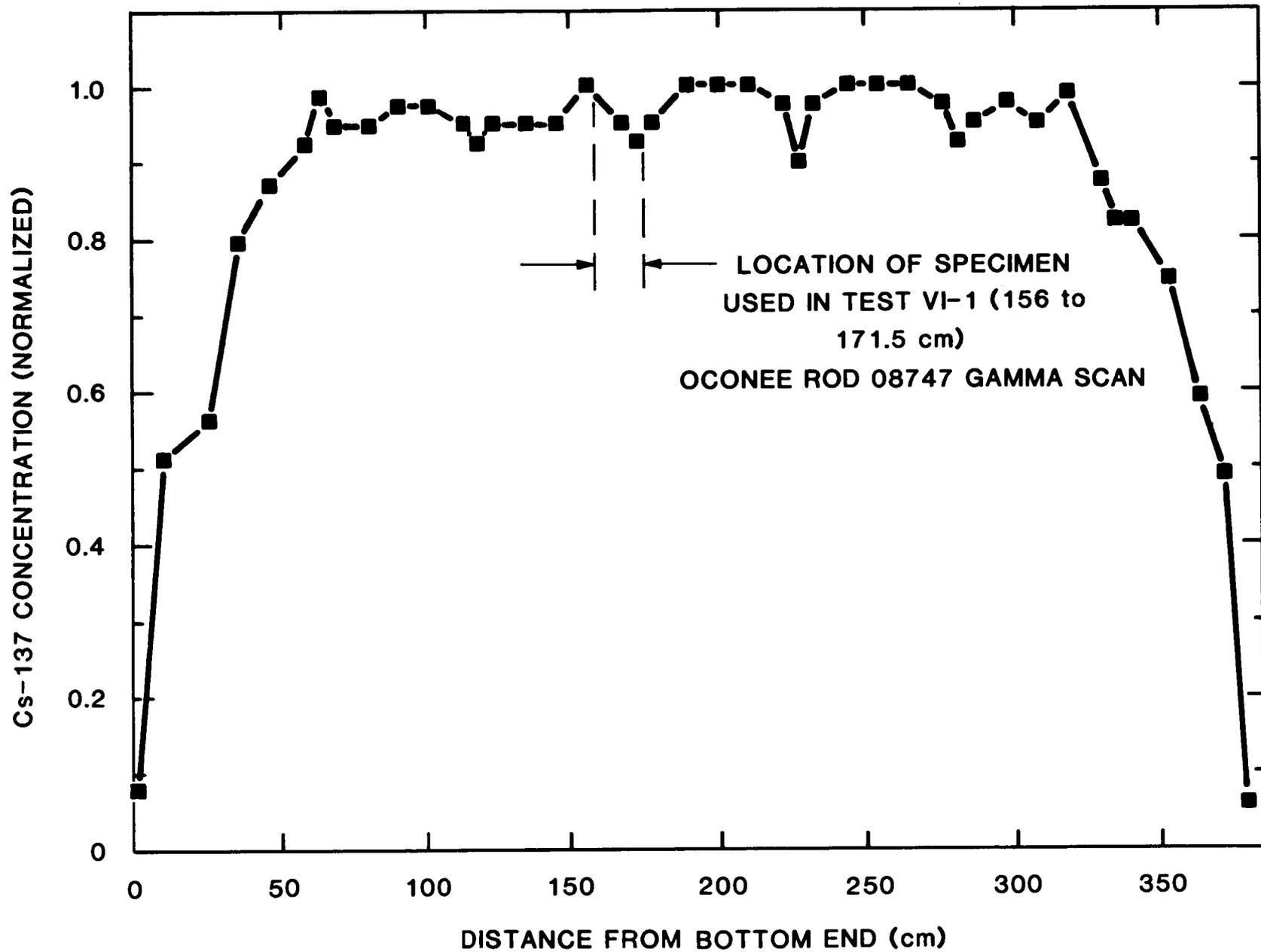


Fig. 1. Axial burnup profile, as indicated by  $^{137}\text{Cs}$ , for Rod 08747, showing location of test VI-1 fuel specimen.

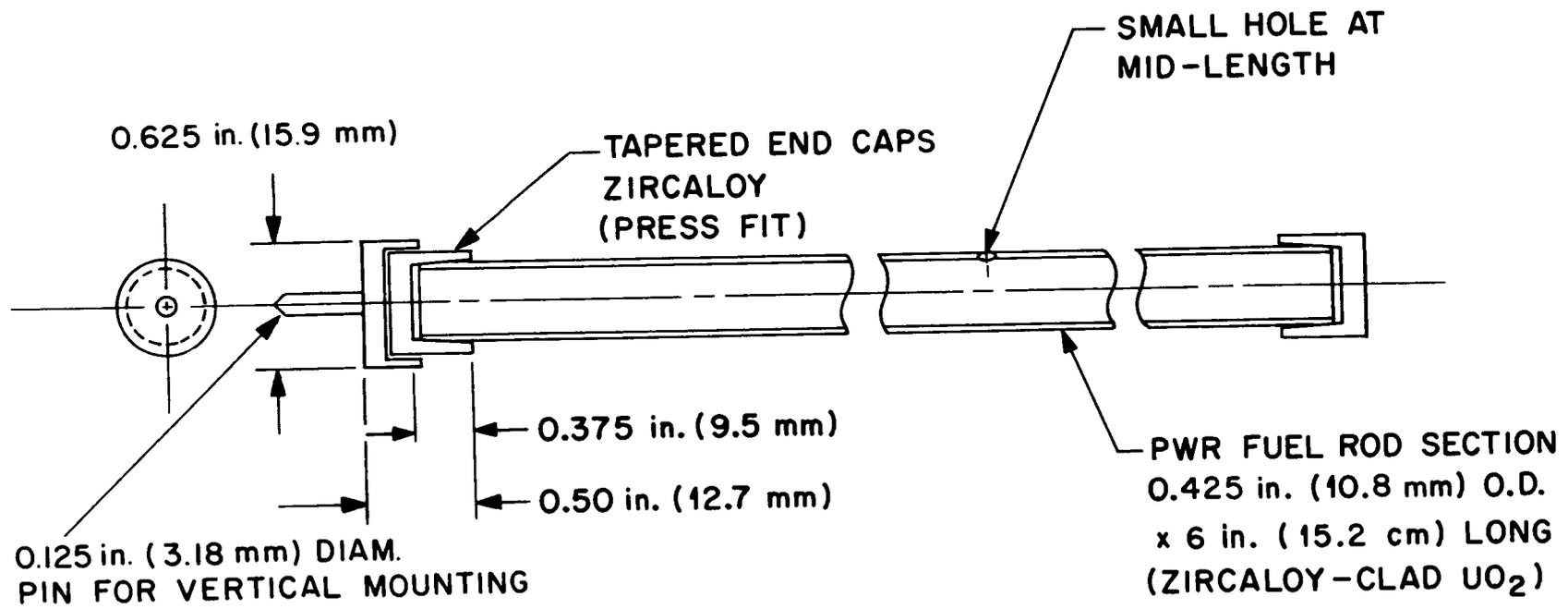


Fig. 2. Dimensions of fuel specimen heated in test VI-1.

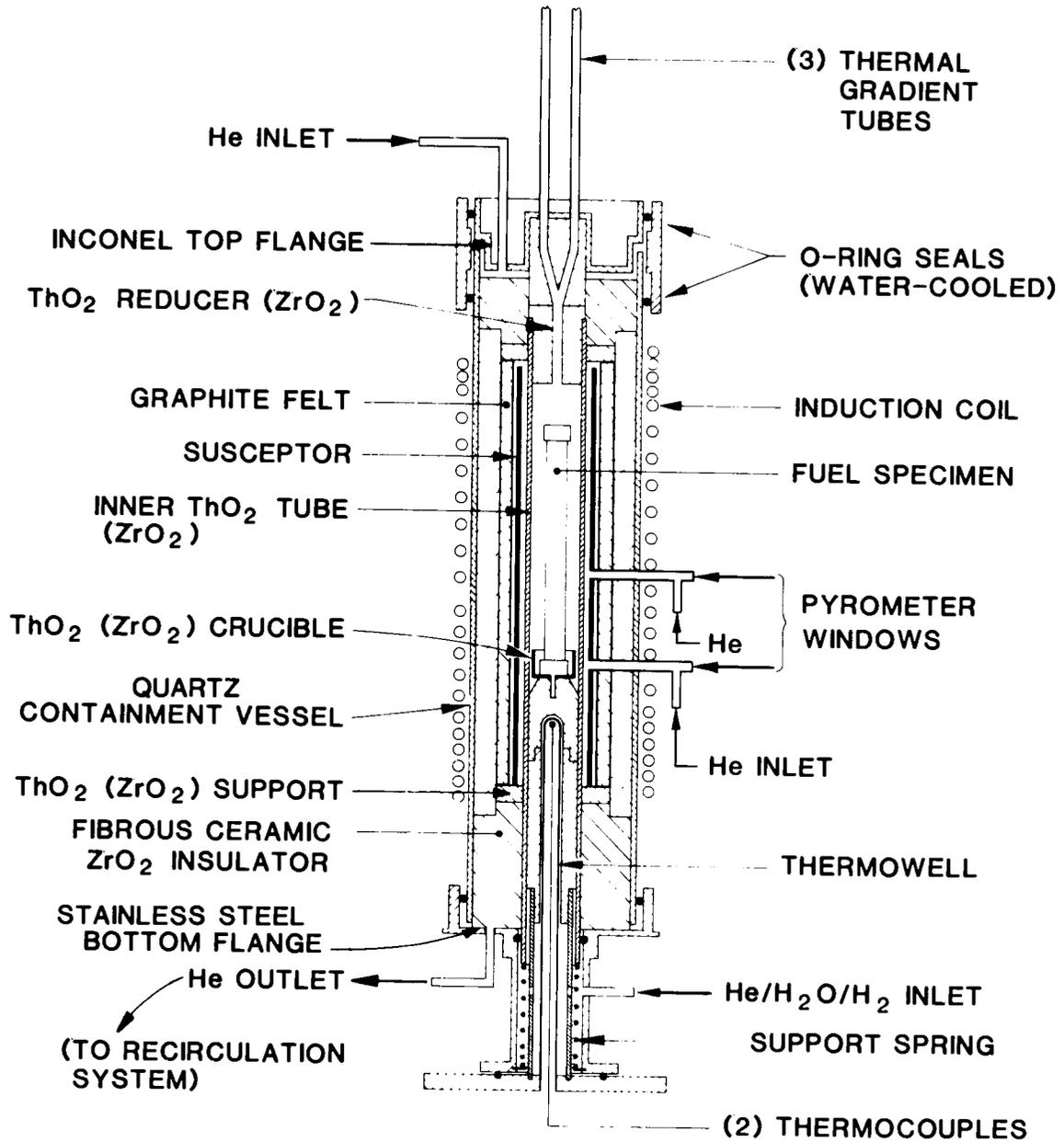


Fig. 3. Details of vertical fission product release furnace.

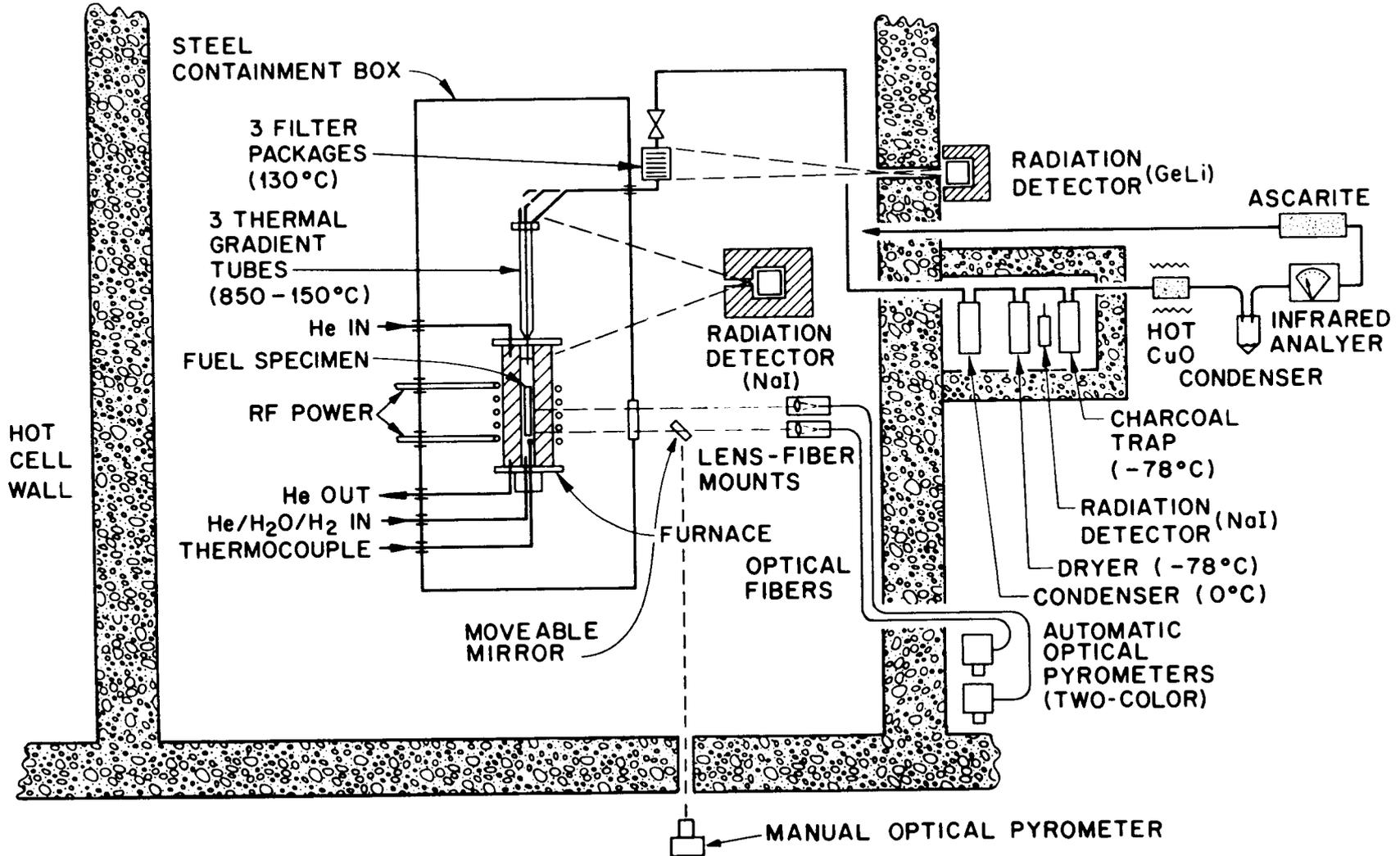


Fig. 4. Vertical fission product release test apparatus.

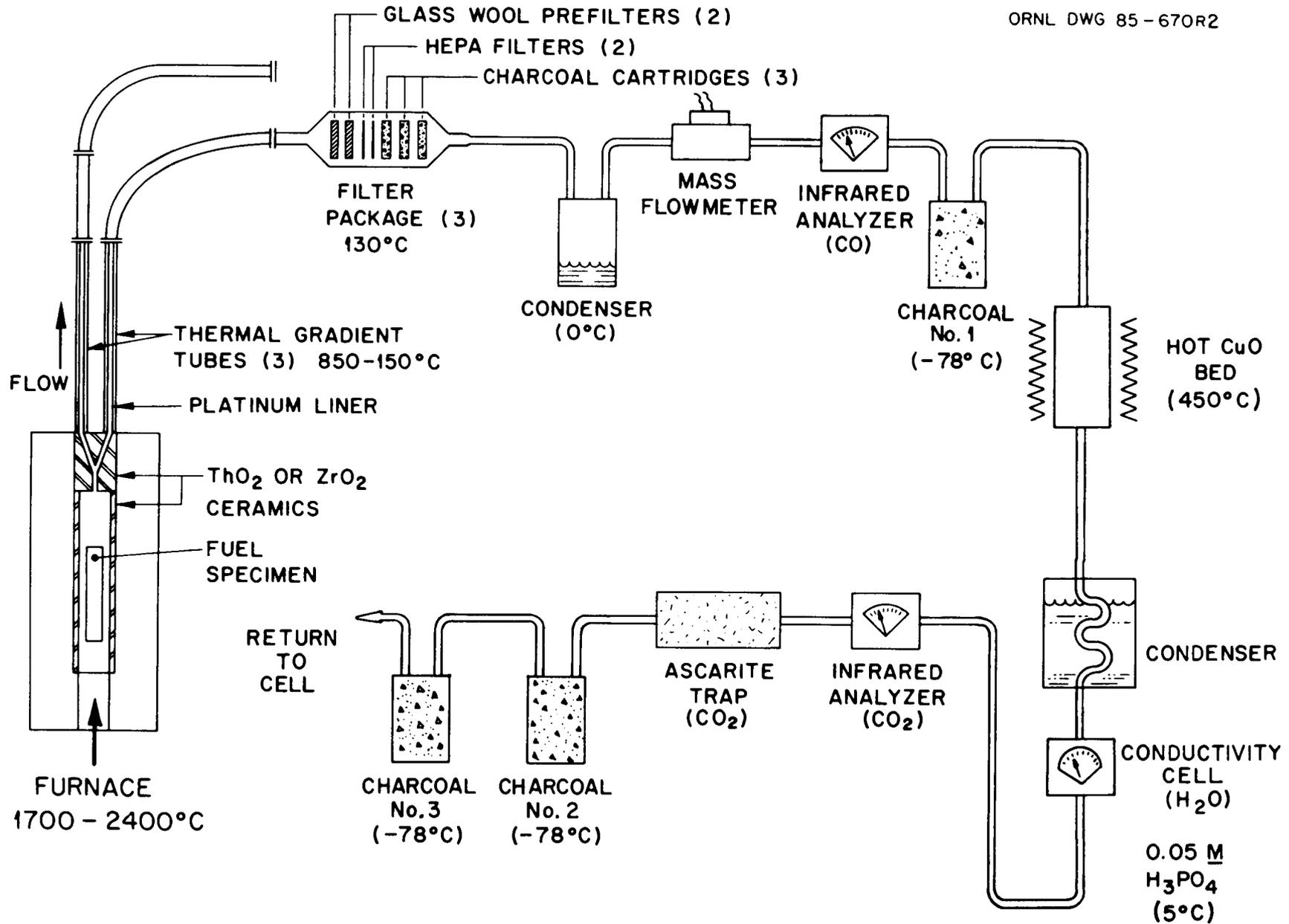


Fig. 5. Components of fission product collection system.

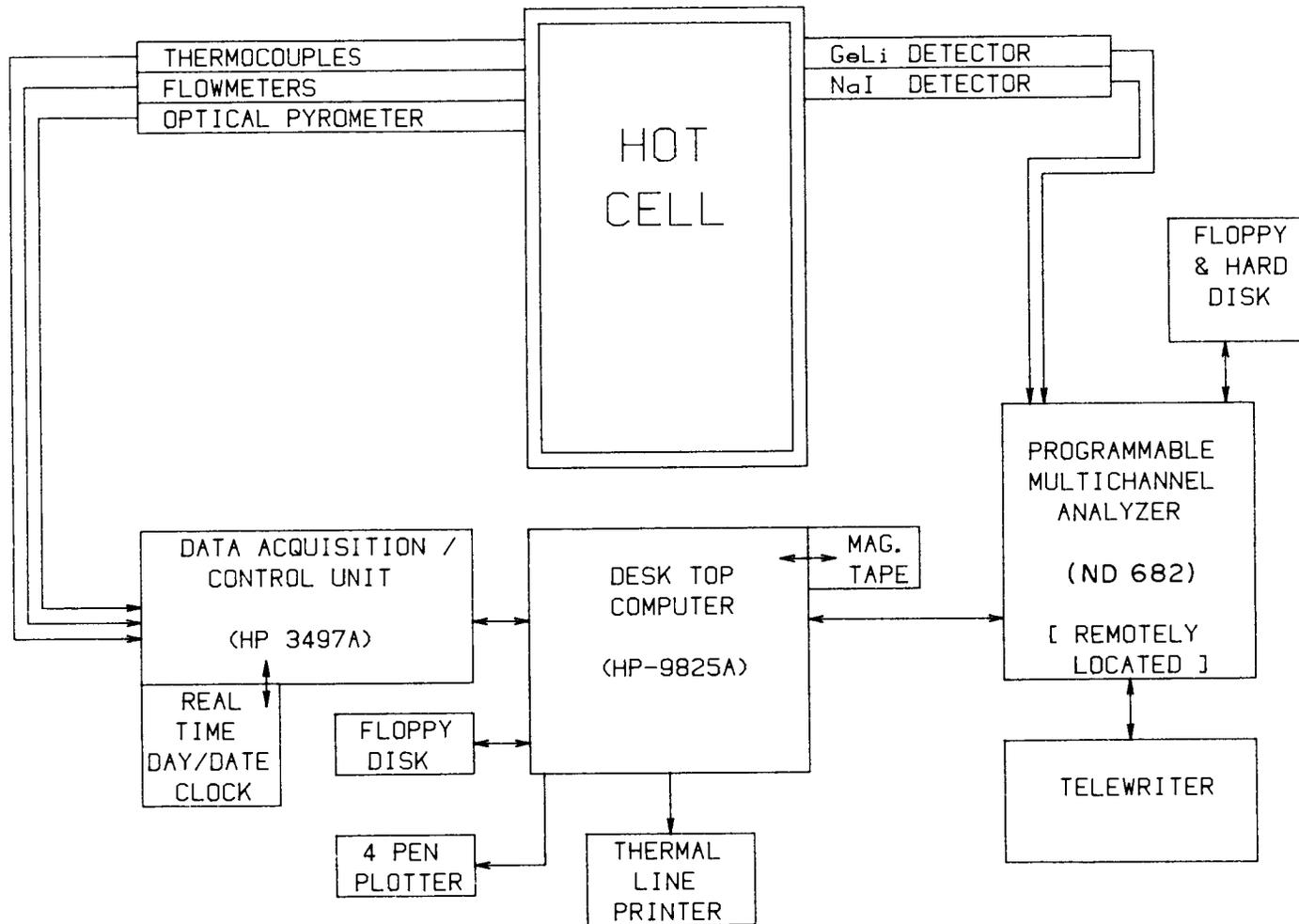


Fig. 6. Data acquisition and processing system for fission product release test.

this test, compared with the previous test in the horizontal apparatus (HI-6), are summarized in Table 6.

Table 6. Summary of apparatus components and test conditions: comparison of tests VI-1 and HI-6

Component/condition	Test VI-1	Test HI-6
Furnace thermocouple	W-5% Re vs W-26% Re (ZrO <sub>2</sub> thermowell)	W-5% Re vs W-26% Re (ZrO <sub>2</sub> thermowell)
Susceptor	Graphite	Graphite
Pretreatment of fibrous ZrO <sub>2</sub> insulator	Zirconyl nitrate (1 coat)	Zirconyl nitrate (1 coat)
Thermal gradient tubes <sup>a</sup> :		
number	3	1
liner material	Platinum	Stainless steel
length (cm)	35.9	37.0
inside diameter (cm)	0.70	0.43
Connector, thermal gradient tube to filter pack	8.0-mm ID × 57- to 89-cm-long stainless steel	4.6-mm ID × 190-mm-long stainless steel
Entrance cone to filter pack	Stainless steel	Stainless steel
Glasswool prefilter	8.9-cm diam with stainless steel screen	5.1-cm diam with stainless steel screen
Dryer and cold charcoal temperature (°C)	-78	-78
Inert gas	Helium	Helium
Aerosol sampler	Stainless steel screw in filter cone	Graphite rod in TGT-filter connec- tor tube

<sup>a</sup>Thermal gradient tube = TGT.

Table 7. Operating data for test VI-1

Specimen temperature (K):	
At start of heatup ramp	~500
During first 20-min period	1410
During second 20-min period	2020
During third 20-min period	2300
Heatup rate	1/s
Cooldown rate	0.8/s
Time above 2000 K	54 min
Nominal gas flow rate data (L/min at 20°C, 1 bar): <sup>a</sup>	
He purge to thermocouple	0.1
He to recirculation system	0.40
He to steam generator	0.30
Steam into furnace	1.54
Recirculation/purification system (He)	1.0
Integral volume flow data (L): <sup>b</sup>	
He purge to thermocouple	15.28
He to recirculation system	52.98
He to steam generator <sup>c</sup>	39.87
Steam into furnace	205
Recirculation	90.8
Total H <sub>2</sub> generated <sup>d</sup>	47.1
H <sub>2</sub> from cladding and end cap oxidation <sup>e</sup>	16.7

<sup>a</sup>Measured by mass flowmeters, except steam measured in condenser.

<sup>b</sup>Measured by totalizers on mass flowmeters during the 133 min of steam flow into apparatus, at room temperature = 24.5°C.

<sup>c</sup>Absolute pressure in furnace during test was 0.09773 MPa (733 mm Hg).

<sup>d</sup>At 1.00 atm = 0.09957 MPa (746.8 mm Hg), measured atmospheric pressure during test.

<sup>e</sup>Theoretical value assuming 100% oxidation of Zr to ZrO<sub>2</sub>.

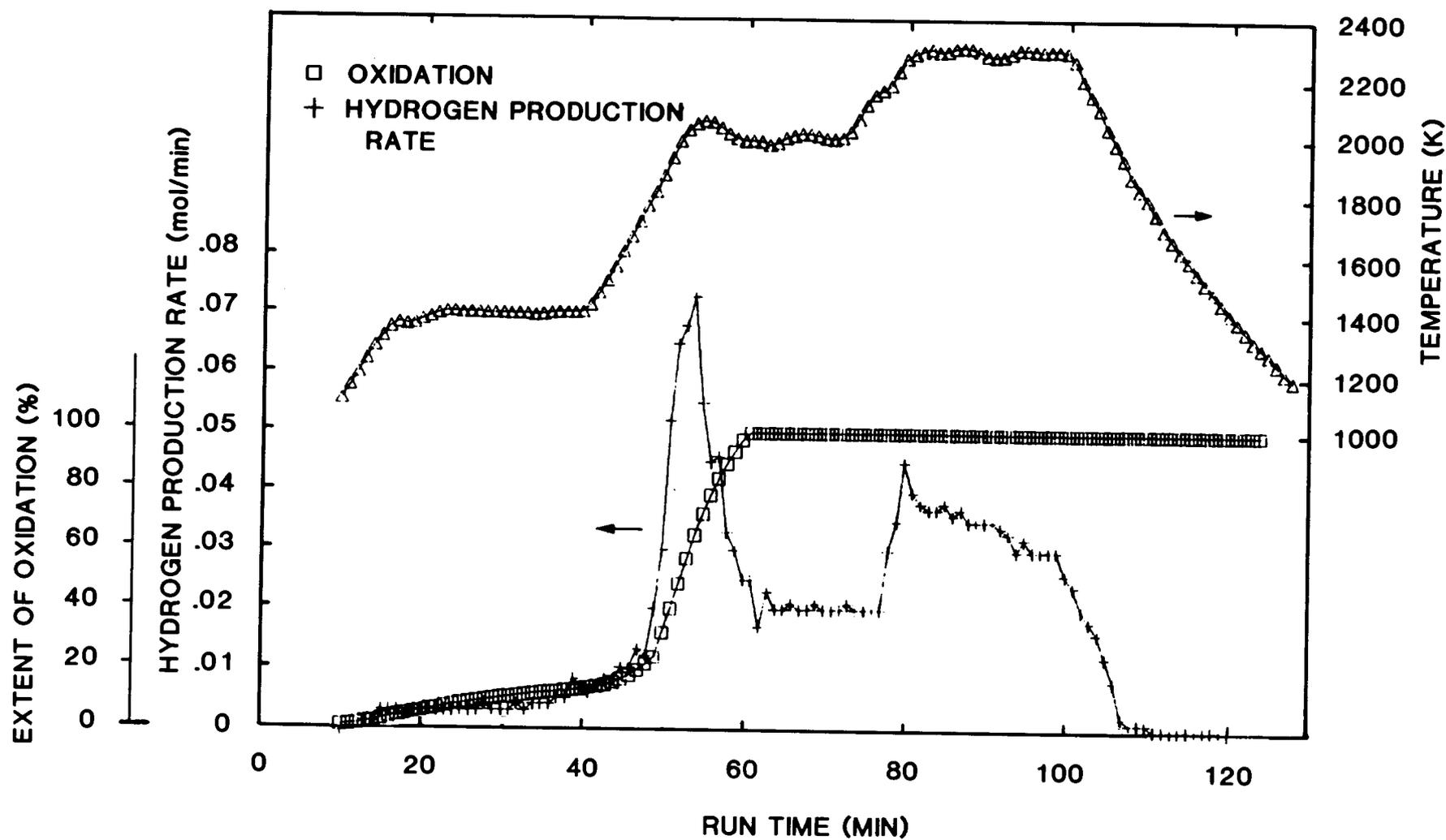


Fig. 7. Cladding oxidation and hydrogen production rate in test VI-1 as a function of time and temperature.

Table 8. Chronology of test VI-1, Dec. 17, 1985

Event/status	Time		Fuel temperature	
	Clock	Test	Pyrometer <sup>a</sup> (°C)	True <sup>b</sup> (K)
Load fuel into furnace	1110			~300 <sup>c</sup>
Begin furnace preheat	1353			~300 <sup>c</sup>
Furnace warm, at low power	1400			~525 <sup>c</sup>
Begin steam flow to furnace	1414			~500 <sup>c</sup>
Test Phase A				
Start ramp	1425	0		~500 <sup>c</sup>
Heating normally	1435	10		~1000 <sup>c</sup>
1st L&N measurement	1436	11	819	1176
Reached first plateau	1445	20	1017	1385
Hold temp. 20 min				
Resumed ramp	1505	40	1043	1413
Reached second plateau	1517	52	1602	2014
Hold 20 min				
Test Phase B				
Switch to Train B, resume ramp	1537	72	1595	2006
Reached third plateau	1545	80	1852	2287
Hold 20 min				
Test Phase C				
Switch to Train C	1551	86	1887	2314
Midpoint	1555	90	1843	2277
Thermocouple failure	1557	92	1848	2282
Power off — end test	1605	100	1862	2297
Down to 2000 K	1610	105	1592	2003
Down to 1400 K	1625	120	1029	1398
Steam off, He only	1627	122	971	1337
Last L&N measurement	1633	128	820	1177

<sup>a</sup>Measured with manually operated L&N optical pyrometer.

<sup>b</sup>Includes corrections for windows, mirror, etc.

<sup>c</sup>Estimated from thermocouple data.

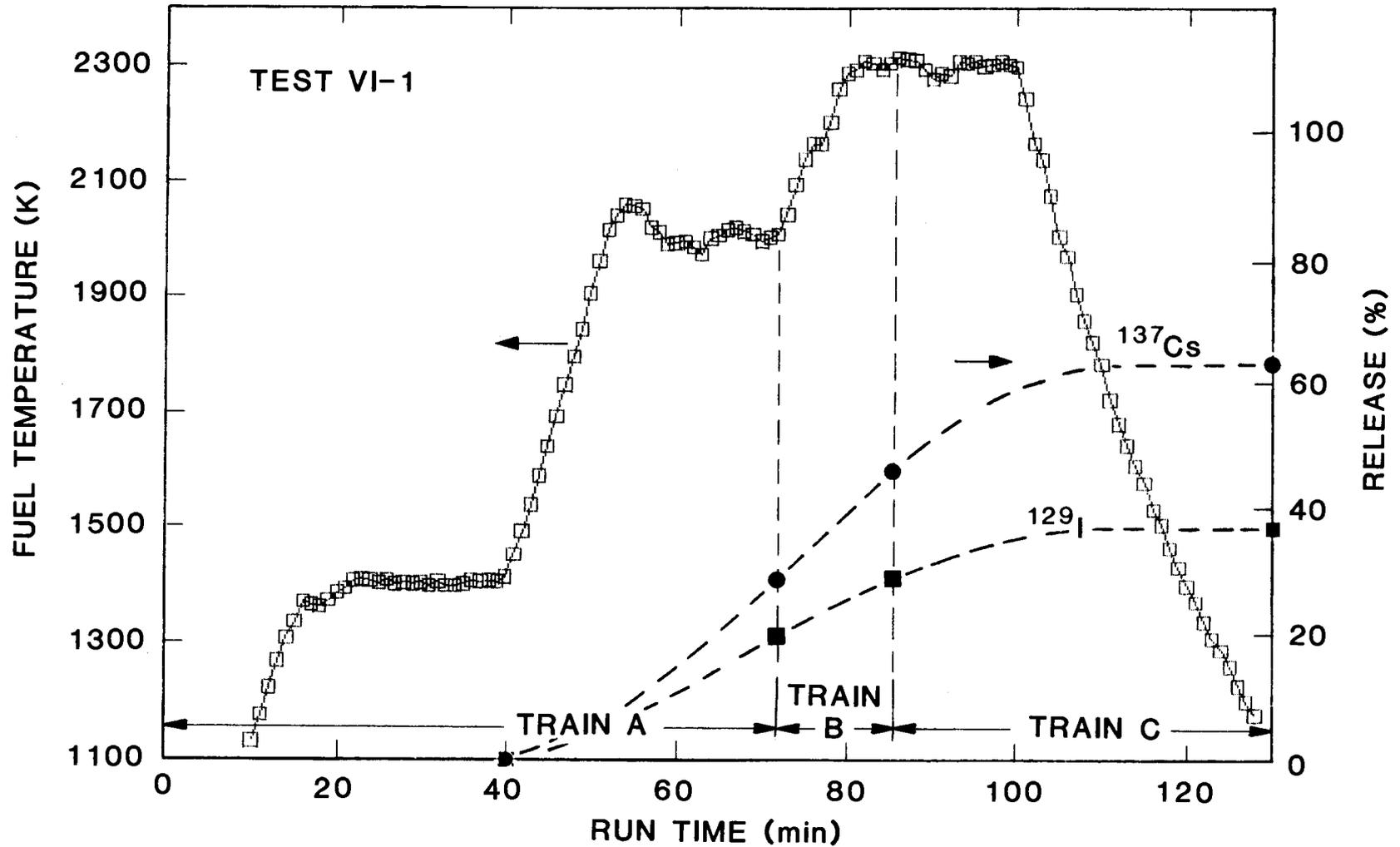


Fig. 8. Temperature and fission product release history in test VI-1.

### 3.3 TEST CONDITIONS AND OPERATION

As in each of the previous experiments, the experimental apparatus was assembled by direct handling, which is possible because the hot-cell and test apparatus are decontaminated after each test. Also, new furnace, TGT, and filter package assemblies are used in each test. Transfer and loading of the highly radioactive fuel specimen and final closure of the furnace and containment box required the use of master slave manipulators. No in-cell operations were required during the test. Before the heating and steam flow was begun, the test apparatus was evacuated and purged with helium. All connecting lines to the furnace-TGT-filter pack assemblies were preheated to at least 125°C to prevent steam condensation during the test.

This test was intended to investigate fission product release at two temperature levels — 2000 and 2300 K — under strongly oxidizing conditions. The operating conditions are summarized in Table 7, and the temperature history is shown in Fig. 7. The more important events during the test are listed in the test chronology, Table 8. The time periods for operation of the three collection trains (Fig. 8) were Train A, 0 to 72 min; Train B, 72 to 86 min; and Train C, 86 min to end of test, including cooldown. (A preheat period was included to slowly heat the specimen to ~550 K prior to beginning steam flow to the furnace. Time zero was defined as that time when the heating ramp was begun, with stable steam flow through the warm furnace.) Temperature measurement and control were generally good; the small peak indicating overshoot at the 2000 K level (~55 min) appears to be (at least partially) the result of oxidation heating, which was near the maximum rate at this time. The 20-min period at 1400 K was included to ensure heatup of ceramics in the outlet end of the furnace and to compare the data from the three optical pyrometers before any significant release of fission products had occurred.

The accumulation of  $^{85}\text{Kr}$  on the cold charcoal traps and of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  on the TGTs and the filters was monitored continuously, as in the HI tests. Unfortunately, higher-than-expected count rates (resulting from changes in the test geometry and cesium penetration of one filter early in the test) caused saturation of the detectors before much on-line release rate data could be obtained. Although the very high count rates from these detectors caused some problems in interpretation during the test, the planned temperature and collection train schedule was followed, and posttest examination of the data and the apparatus provided a thorough understanding of the test events.

### 3.4 POSTTEST DISASSEMBLY AND EXAMINATION

After the test was completed, the apparatus was monitored for the distribution of radioactivity and then disassembled. Initially, the filter assemblies and the TGT liners were removed and transferred to another hot cell to avoid potential contamination from fuel handling. The top flange was removed from the furnace, and after removal of the ceramic components from the outlet end, the top end of the fuel specimen

could be observed, as shown in Fig. 9. The Zircaloy end cap appeared to be heavily oxidized, and the top end of the specimen was tilted to make contact with the  $ZrO_2$  furnace tube, but the specimen remained largely intact and upright. To preserve this geometry for sectioning and subsequent microstructural examination, the furnace tube was filled with epoxy resin. After the resin had hardened, the furnace tube/fuel specimen assembly was transferred to the High Radiation Level Examination Laboratory for the examination of cross sections.

#### 4. TEST RESULTS

##### 4.1 TEST DATA

Test temperature is compared with the hydrogen production rate and the extent of cladding oxidation in Fig. 7. Hydrogen was measured by conversion of  $H_2$  to  $H_2O$  in the hot  $CuO$  bed, followed by collection of the  $H_2O$  in the condenser, as indicated in Fig. 5. The production rate curve in Fig. 7 indicates a peak at the beginning of the 2000 K period, then a decline as most of the Zircaloy cladding became fully oxidized to  $ZrO_2$ . The second rate increase (at ~79 min) coincides with the temperature increase to 2300 K. Continued hydrogen production at a declining rate resulted from oxidation of (1) the thicker Zircaloy end caps, (2) the  $UO_2$  fuel, and (3) the graphite susceptor, by steam leaking into that region. This oxidation behavior, as indicated by hydrogen production, agreed quite well with the model developed by Yamashita<sup>15</sup> (see Fig. 10). The measured oxidation rate at 1700 K was slightly below that predicted by the model using oxidation rate data from Baker and Just,<sup>16</sup> and, at 2000 K, the measured rate slightly exceeded the rate predicted by the model using rate data from Urbanic and Heidrick.<sup>17</sup> The overall agreement, however, between the measured rates and those predicted by the model using data from these recognized sources is quite good.

##### 4.2 POSTTEST DATA

###### 4.2.1 Gamma Spectrometry

As in previous tests, all experimental components and collectors were analyzed after the test under well-defined geometry to determine the content of the gamma-emitting fission products. In addition, a new technique was developed for processing the data to provide more precise results. Because all samples required at least double containment (metal, glass, or plastic) and because many samples were actually distributed within other materials (especially the fuel specimen itself), it was difficult to calculate accurately the effective shielding for the various gamma-ray energies of interest. Considering the ubiquitous nature of cesium and the broad range of gamma-ray energies inherent to  $^{134}Cs$  (nine major lines at 475 to 1365 keV), an empirical method of determining the effective shielding to obtain a mass balance for cesium among several of the  $^{134}Cs$  gamma-ray energies was developed. This shielding value was then applied to other nuclides, such as  $^{106}Ru$ ,  $^{110m}Ag$ , and  $^{125}Sb$ , to provide more accurate results.

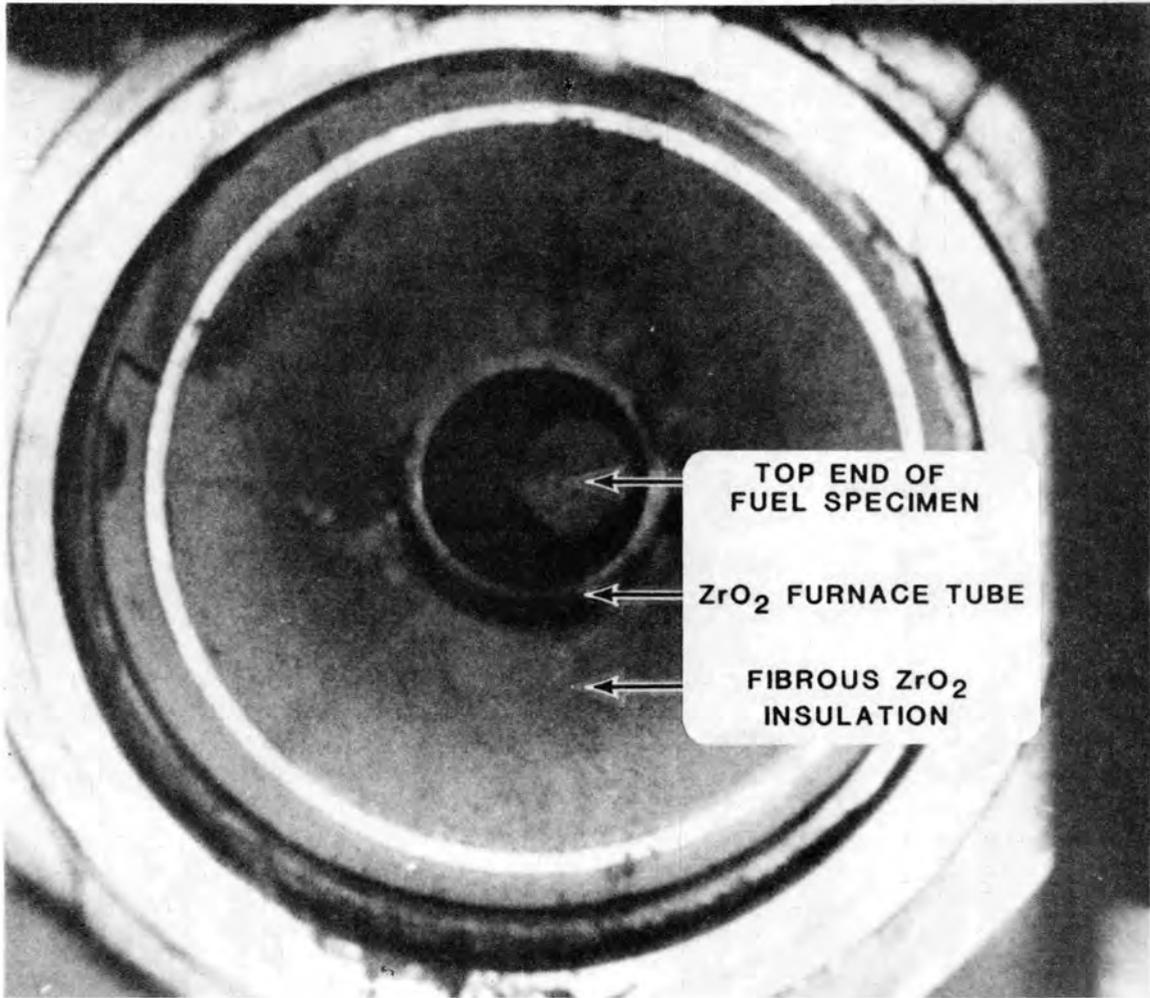


Fig. 9. Top view of fuel inside furnace, test VI-1.

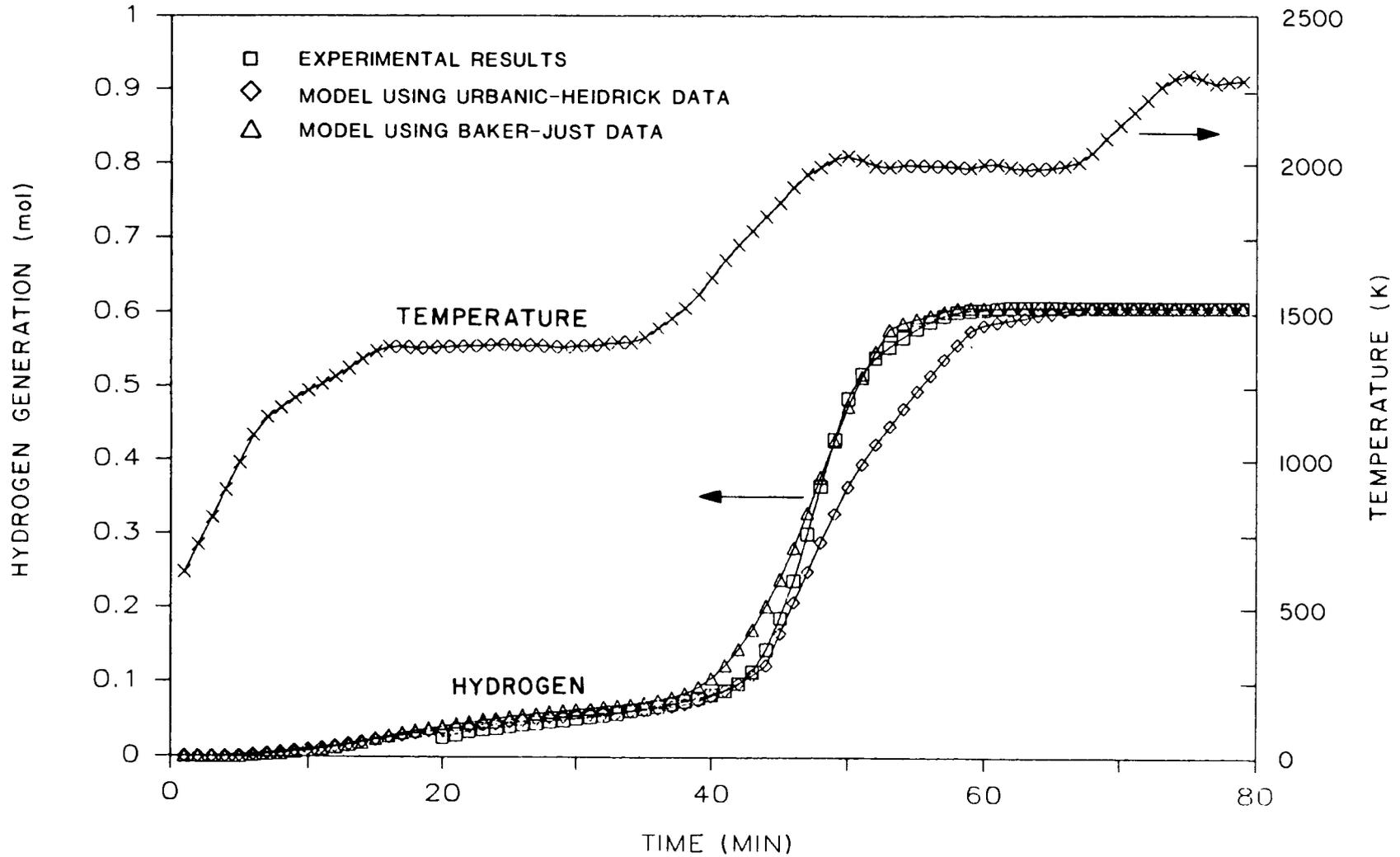


Fig. 10. Comparison of measured and calculated hydrogen production in a test simulating VI-1.

Pretest gamma spectrometric analysis of the fuel specimen (a 15.2-cm-long section cut from rod 08747 from the Oconee Unit 1 reactor,<sup>13</sup> as described previously) was used to determine the fission product inventories in the fuel. Long-lived gamma emitters —  $^{106}\text{Ru}$ ,  $^{125}\text{Sb}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{154}\text{Eu}$  — were counted directly; a calculation by the computer program ORIGEN 2, with corrections based on  $^{137}\text{Cs}$ , supplied the inventory values for other fission products, activation products, and fuel nuclides, as shown in Tables 3 and 4. The distributions of the fission products measured in the fuel before and after the test are shown in Fig. 11. The pretest curves, based on measurements at 2-cm intervals, indicate that some migration of cesium and antimony away from the outlet end of the specimen had occurred during irradiation, whereas the less volatile elements, ruthenium and europium, remained stationary in the  $\text{UO}_2$  and exhibited relatively flat profiles.

As has been typical of these tests,  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  were the dominant activities in almost all samples, and interfered with the analysis of less abundant fission products. A summary of the fractional release results, as determined by gamma spectrometry, and of  $^{129}\text{I}$ , which is measured by leaching and neutron activation analysis, is presented in Table 9. The total release values for  $^{85}\text{Kr}$  and  $^{137}\text{Cs}$  were in reasonably good agreement, 47 and 63% of fuel inventory respectively. The krypton release value includes an estimated 0.7% of inventory that was released from the  $\text{UO}_2$  to the pellet-cladding gap during irradiation, as indicated by gas collection and measurement prior to sectioning of the fuel.<sup>13</sup> Although no data for  $^{134}\text{Cs}$  are shown, the agreement between  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  was quite good; the total release of  $^{134}\text{Cs}$  was 65%. As has occurred frequently in previous tests, the indicated release for  $^{129}\text{I}$  was significantly less (37%) and will be discussed later. The indicated release values for  $^{110\text{m}}\text{Ag}$  and  $^{125}\text{Sb}$ , which are fission products of much lower yield and are generally less hazardous, are higher than have been observed previously. The fully oxidized cladding, a condition not existing in previous tests at  $\sim 2300$  K, is suspected of being significant in these higher release values. It should be noted that large fractions ( $\sim 1/3$ ) of the released Ag and Sb were deposited on  $\text{ZrO}_2$  surfaces in the outlet region of the furnace, where temperatures were 1100 to 1500 K.

The detailed distribution of  $^{137}\text{Cs}$  throughout the test apparatus is shown in Table 10, and the values are converted to total mass of cesium, based on ORIGEN2 data. The distributions of  $^{110\text{m}}\text{Ag}$  and  $^{125}\text{Sb}$  are summarized in Table 11. Fractional release values are based on the total amount of nuclide found after the test. The low levels of  $^{110\text{m}}\text{Ag}$  were not readily measurable in the fuel, but could be measured in the collectors where there was less self-shielding and interference from other nuclides, most notably  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ , and  $^{154}\text{Eu}$ .

#### 4.2.2 Neutron Activation Analysis for Iodine

Since iodine has no long-lived, gamma-emitting nuclides, analytical methods other than gamma spectrometry must be used. Neutron activation of  $^{129}\text{I}$  to  $^{130}\text{I}$ , which can be counted easily, is a proven, sensitive technique. Iodine forms dissolve readily in basic solutions to form

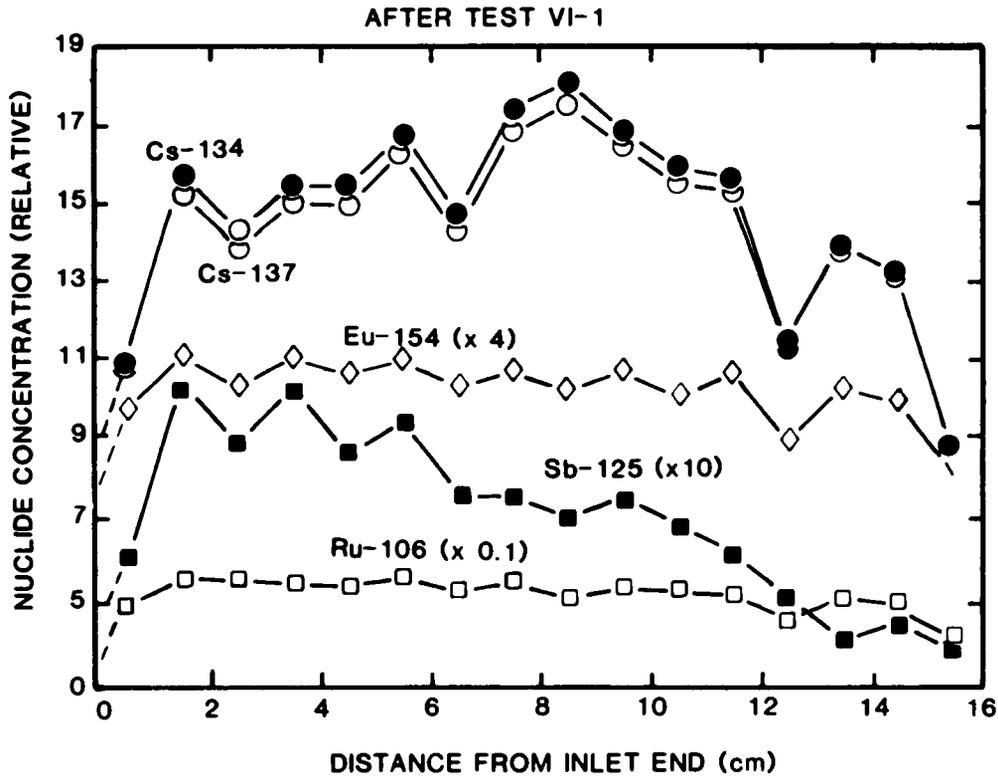
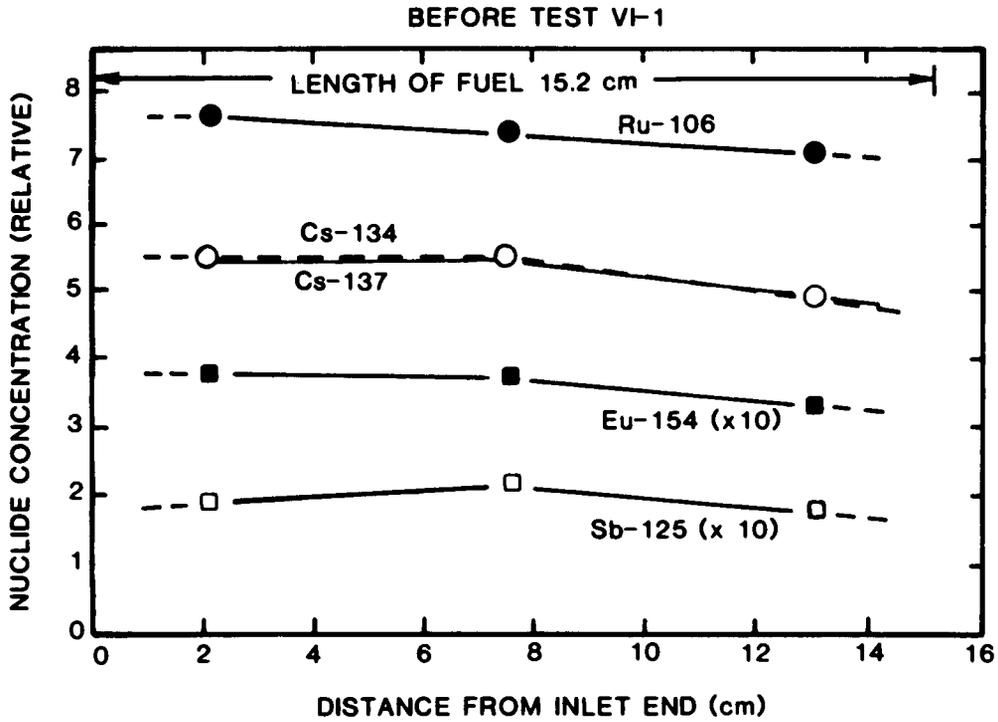


Fig. 11. Pretest and posttest distributions of fission products in test VI-1 fuel specimen.

Table 9. Summary of release data for test VI-1

Component/collector	Percent of fission product inventory <sup>a</sup> found				
	<sup>85</sup> Kr	<sup>110m</sup> Ag	<sup>125</sup> Sb	<sup>129</sup> I	<sup>137</sup> Cs
Furnace	0	30.01	10.15	0 <sup>b</sup>	4.00
Train A					
TGT		7.36	0.40	4.09	11.50
Filters <sup>c</sup>		1.61	0	13.90	15.06
Hot charcoal		0	0	0.06	0
Total	0	8.97	0.40	19.05	26.91
Train B					
TGT		3.65	2.40	2.37	6.12
Filters <sup>c</sup>		6.49	4.94	6.86	10.28
Hot charcoal		0	0	0.14	0
Total		10.14	7.34	9.36	16.40
Train C					
TGT		17.64	9.12	2.10	6.85
Filters <sup>c</sup>		16.75	6.38	6.57	9.02
Hot charcoal		0	0	0.04	0
Total		34.43	15.50	8.71	15.87
Cold charcoal	46.0	0	0	0	0.05
Total release	46.7 <sup>d</sup>	83.55	33.38	37.12	63.23

<sup>a</sup>Inventories based on pretest and posttest fuel analysis by gamma spectrometry; ORIGEN2 calculations were used to determine inventories of nuclides not gamma-measurable.

<sup>b</sup>Not all of furnace was leached and analyzed for <sup>129</sup>I; furnace thermal insulation probably contained small amounts.

<sup>c</sup>Includes deposits in lines to filters.

<sup>d</sup>Includes an estimated 0.7% released during irradiation.<sup>14</sup>

Table 10. Fractional release and distribution of cesium in test VI-1<sup>a</sup>

Location	Approximate temperature (K)	Cesium found at each location			
		<sup>137</sup> Cs (μCi)	Total Cs (mg)	(Percent of inventory)	(Percent of released)
Furnace components					
First ZrO <sub>2</sub> plug	~2000	7356	0.194	0.056	0.09
Second ZrO <sub>2</sub> plug	~1500	402500	10.634	3.065	4.85
ZrO <sub>2</sub> donut	500-1200	104100	2.750	0.793	1.25
Exit flange	350-1150	2540	0.067	0.019	0.03
Inlet parts	400-1500	8794	0.232	0.067	0.11
Total		525290	13.878	4.001	6.33
Train A					
TGT A	420-1160	1510000	39.894	11.500	18.19
TGT - filter line	430	202300	5.345	1.541	2.44
First prefilter	400	1395000	36.856	10.625	16.80
Second prefilter	400	388200	10.256	2.957	4.68
HEPA filters	400	38330	1.013	0.292	0.46
Total		3533830	93.364	26.914	42.57
Train B					
TGT B	420-1170	803900	21.239	6.123	9.68
TGT - filter line	430	162600	4.296	1.238	1.96
First prefilter	400	980800	25.913	7.470	11.81
Second prefilter	400	187400	4.951	1.427	2.26
HEPA filters	400	18890	0.499	0.144	0.23
Total		2153590	56.898	16.402	25.94
Train C					
TGT C	420-1200	899300	23.760	6.849	10.83
TGT - filter line	430	141100	3.728	1.075	1.70
First prefilter	410	894500	23.633	6.813	10.77
Second prefilter	410	134600	3.556	1.025	1.62
HEPA filters	410	13450	0.355	0.103	0.16
Total		2082950	55.032	15.864	25.09
Total in condenser and cold charcoal		5851	0.155	0.045	0.07
Total found outside fuel		8301511	219.326	63.226	99.99
Total in fuel (after test)		4846000	102.031	42.7 <sup>b</sup>	

<sup>a</sup>Inventory based on measured data, 13.13 Ci <sup>137</sup>Cs on July 15, 1981.

<sup>b</sup>Total posttest counts indicate 106% of pretest counts.

Table 11. Fractional release and distribution of silver and antimony in test VI-1<sup>a</sup>

Location	<sup>110m</sup> Ag		<sup>125</sup> Sb	
	( $\mu$ Ci)	(% inventory)	( $\mu$ Ci)	(% inventory)
Furnace components				
First ZrO <sub>2</sub> plug	0	0	1657	0.33
Second ZrO <sub>2</sub> plug	21884	28.79	45430	9.07
ZrO <sub>2</sub> donut	909	1.20	3468	0.69
Other	11	0.01	305	0.06
Total	22804	30.01	50860	10.15
Train A				
TGT A	5590	7.36	2000	0.40
TGT - filter line	122	0.16	17	0.00
First prefilter	1000	1.32	0	0.00
Second prefilter	0	0.00	0	0.00
HEPA filters	100	0.13	0	0.00
Total	6812	8.96	2017	0.40
Train B				
TGT B	2775	3.65	12000	2.40
TGT - filter line	500	0.66	2844	0.57
First prefilter	4433	5.83	19380	3.87
Second prefilter	0	0.00	2175	0.43
HEPA filters	0	0.00	337	0.07
Total	7708	10.14	36736	7.33
Train C				
TGT C	13410	17.64	45700	9.12
TGT - filter line	1533	2.02	5056	1.01
First prefilter	9229	12.14	22450	4.48
Second prefilter	2000	2.63	4035	0.81
HEPA filters	0	0.00	382	0.08
Total	26172	34.44	77623	15.49
Total outside fuel	63496	83.55	167236	33.38
Total in fuel (after test)	N.D. <sup>b</sup>		331000	66.07

<sup>a</sup>Inventories based on measured data: 501 mCi <sup>125</sup>Sb and 76 mCi <sup>110m</sup>Ag on July 15, 1981.

<sup>b</sup>N.D. = not detected.

stable iodides; in our samples, large amounts of highly radioactive cesium were also dissolved in the  $\text{NH}_4\text{OH} + \text{H}_2\text{O}_2$  leach solution. Small aliquots of the solutions were chemically treated to remove cesium, loaded onto ion-exchange media, irradiated, and the 12.5-h  $^{130}\text{I}$  was counted. The results of these analyses, along with data for fractional iodine release and cesium/iodine ratios at various apparatus locations, are summarized in Table 12. The total fraction of iodine released was a minimum value. Since the measurement of  $^{129}\text{I}$  involves a series of steps, each of which provides an opportunity for a loss of iodine, we believe that the true release value is significantly higher, probably similar to the Kr and Cs values. The important observations from the iodine distribution data are (1) ~25% of the iodine in each train was deposited in the TGT and almost all of the remainder on the filters and (2) more than half of the released iodine was found in Train A, as was the case for cesium. These distribution patterns are consistent with those of previous tests under similar conditions. In the first observation, the larger fraction of the released iodine, normally 65 to 75%, has been consistently found on the filters, indicating that it was associated with the aerosol. As always, only a tiny fraction (~0.2% in this test) was of a volatile form that could penetrate the filters to be collected on the charcoal or in the condenser. In the second observation, the iodine, like cesium, was released predominantly at the lower temperatures (2020 K) of phase A, and the source likely was depleted to the extent that, even though the release rate was higher at the higher temperature (2300 K) of phase C, the indicated total release fraction during this period (7.7%) was much smaller than that during phase A (17%). The Cs/I mass ratio column in Table 12 shows that the ratio declined with distance through the collection train. This behavior of cesium and iodine is consistent with previous experiments and with our understanding of the chemical species involved: most of the iodine appears to be in the form CsI, which is stable and unreactive; whereas most of the cesium is in the highly reactive form CsOH and tends to react quickly, thereby becoming rapidly depleted as it passes through the TGTs and prefilters.

#### 4.2.3 Spark Source Mass Spectrometry

Small smear samples of the deposits on the three TGTs were collected on graphite electrodes and were analyzed by spark source mass spectrometry (SSMS). Gamma scans of the TGTs, which showed the distributions of  $^{137}\text{Cs}$  and  $^{125}\text{Sb}$ , were used to determine the sample locations. Assuming that the smear samples were representative of the deposits at the selected locations, the easily measured  $^{137}\text{Cs}$  (by gamma spectrometry) was used to quantify the relative amounts of the other elements — fission products, structural materials and impurities — vaporized in the mass spectrometer. This technique has the advantage of providing data for a wide variety of elements, most of which are not radioactive and are not readily measured otherwise.

These data are summarized in Table 13. In addition to cesium, the fission product elements selenium, bromine, rubidium, molybdenum, silver, cadmium, tellurium, iodine, and barium were detected in one or more samples. The deposits from the cooler regions of TGTs B and C contained

Table 12. Fractional release and distribution of iodine in test VI-1

Component/collector	Mass of I ( $\mu\text{g}$ )		Percent of inventory <sup>a</sup>	Cs/I mass ratio
	<sup>129</sup> I	Total I		
Train A				
TGT A	1069	1410	5.09	28.3
Prefilters <sup>b</sup>	2770	3654	13.19	14.4
HEPA filters	150	198	0.71	5.12
Charcoal	11.1	14.6	0.05	
Condenser	2.0	2.6	0.01	
Total train A	4002	5279	19.05	17.7
Train B				
TGT B	497	656	2.37	32.4
Prefilters <sup>b</sup>	1365	1800	6.50	19.5
HEPA filters	74	98	0.35	5.1
Charcoal	26.5	35	0.13	
Condenser	1.9	2.5	0.01	
Total train B	1964	2592	9.36	21.9
Train C				
TGT C	440	580	2.10	41.0
Prefilters <sup>b</sup>	1328	1752	6.23	17.6
HEPA filters	53	70	0.25	5.1
Charcoal	5.3	7.0	0.03	
Condenser	1.7	2.2	0.01	
Total train C	1828	2411	8.71	22.8
Total for test	7794	10282	37.12	21.3

<sup>a</sup>Assumes 21.00 mg <sup>129</sup>I inventory in fuel, based on ORIGEN2 calculation.

<sup>b</sup>Includes iodine from TGT exit block, connecting lines, and filter entrance cones, partitioned according to <sup>137</sup>Cs.

Table 13. Distribution of fission products and other materials in test VI-1  
(spark source mass spectrometry data, wt %)

Train Position (cm) <sup>a</sup>	A 11	A 16	A Filter	B 3	B 11	B 34	B Filter	C 6	C 26	C 35
Fission products										
Se			0.4				0.7			
Br			4.4				0.0 <sup>b</sup>			
Rb	8.9	4.0	14.6	1.0	0.8	6.1	0.6	0.3	3.0	2.1
Mo			20.3			14.4	0.6	0.0	0.0	26.4
Ag			14.6				0.5		0.0	4.7
Cd			1.5				2.8		13.7	0.0
Te			1.5			28.6	1.4	0.0	40.8	7.4
I			3.7		0.0	5.5	0.0	0.0	14.5	0.0
Cs	22.6	10.4	3.4	3.6	2.9	20.0	1.5	1.2	10.9	4.6
Ba			0.0		0.0	0.3	0.1	0.0	0.0	0.0
Reactor materials										
Zr			3.8				2.8			
Sn			0.0	0.0	0.0	16.9	45.4	0.0	0.0	28.9
U			0.0				37.4			
B	0.0	0.1	0.4	0.1	0.0	0.0	0.0	0.0	0.0	0.0
Fe	3.4	13.2	7.3	3.8	0.0	1.3	0.9	2.5	5.7	4.0
Cr	0.0		2.9	0.0		0.0	0.5		0.0	0.0
Ni	0.0		0.7				0.2		0.0	
Co			0.0				0.2		0.0	
Mn			0.1				0.5			
Cu	0.0	0.0	1.5				0.2		0.0	0.0
Other <sup>c</sup>										
Na	2.7	2.7	4.4	2.8	3.9	1.4	1.7	1.7	1.4	1.3
Al		0.0	1.5	3.6	2.7	0.6	0.0	2.5	0.8	0.6
Cl			1.5			0.0	0.5			0.0
Mg			0.2				0.0			
Si			1.5				0.5			
P		1.0	0.3	1.3	0.0	0.7	0.1	0.9	1.2	0.8
S	55.4	53.2	0.4	68.8	79.9	3.4	0.9	81.1	2.0	4.2
K		2.2	4.4	1.6	0.0	0.0	0.5	0.7	0.0	0.6
Ca	7.0	13.3	4.4	13.5	9.9	0.9	0.5	9.1	6.1	14.5
Zn			0.4			0.0	0.1			
V			0.0				0.5			
Sum	100.0	100.0	100.1	100.0	100.0	100.0	100.0	100.0	100.0	

<sup>a</sup>Samples were taken at selected locations along the TGTs.

<sup>b</sup>Zero (0.0) values indicate <0.05 wt %.

<sup>c</sup>Structural materials and impurities from ceramics.

significant fractions of Mo and Te — 20 to 40 wt %. The only previous test in which comparable amounts of Mo and Te were found was test HI-2, the only test in the HI series with completely oxidized cladding. This behavior of tellurium (i.e., minimal release as long as metallic Zircaloy is available for retention) is consistent with previous work.<sup>18</sup>

As indicated by gamma analyses (Sect. 4.2.1), the highly volatile cesium (and its analog rubidium) was found in greatest concentrations in the deposits from TGT A. In addition, large fractions of sulfur, an impurity in the ZrO<sub>2</sub> furnace ceramics, were found at most TGT locations. Pretest treatment procedures designed to remove most of this sulfur have been used successfully in subsequent tests.

Samples of the acid leach solutions from the filters, which were located downstream from the filters, were analyzed also. Unfortunately, no results were obtained from the C train filter because of insufficient material. The A and B filters exhibited a broader range of elements (both fission products and other materials) than did the TGT samples. Particularly large concentrations of Rb, Mo, and Ag were found on filter A. Some 37% of the material on filter B was uranium, but no uranium was found by SSMS at any other location. Additional and more precise data for uranium were obtained by fluorimetric analysis, as reported in Sect. 4.2.5.

#### 4.2.4 Thermal Gradient Tube Deposits

The <sup>137</sup>Cs profiles and the temperature profiles for the period of collection in the three TGTs are shown in Figs. 12 to 14. In addition, small amounts of <sup>125</sup>Sb were found in all three tubes, especially near the inlet ends. It should be noted (see Table 9) that the largest amounts of <sup>137</sup>Cs were found in TGT A (Fig. 12), and that only this tube exhibited the "characteristic" cesium peak (as normally observed in previous tests) at 773 to 873 K (500 to 600°C). Small <sup>137</sup>Cs peaks are apparent near the outlet end of all three tubes, possibly resulting from flow disturbances at the transition connection. The volume gas flow rate in this test was approximately twice that used in most of the HI series tests. Consequently, some differences in location and shapes of deposit peaks would be expected. A large <sup>137</sup>Cs peak occurred in the high temperature region near the inlet of TGT C (Fig. 14).

Earlier work indicated that, under these test conditions, most of the fission product iodine would combine with cesium, forming CsI, and that most of the remaining cesium would be in the form CsOH. In our TGT, such compounds should deposit in the temperature range 500 to 600°C, as observed in Fig. 12. The high temperature peak in TGT C (Fig. 14) suggests the presence of a form of cesium having a deposition temperature of ~1023 K (~750°C), but no identification has been possible. The limited data for <sup>125</sup>Sb indicate that the deposits near the TGT inlets were of the elemental form, as suggested by Norwood, whereas the small amounts of <sup>125</sup>Sb found throughout the TGT (Fig. 14) were associated with, and deposited with, aerosol particles.<sup>19</sup> Unfortunately, no antimony (a

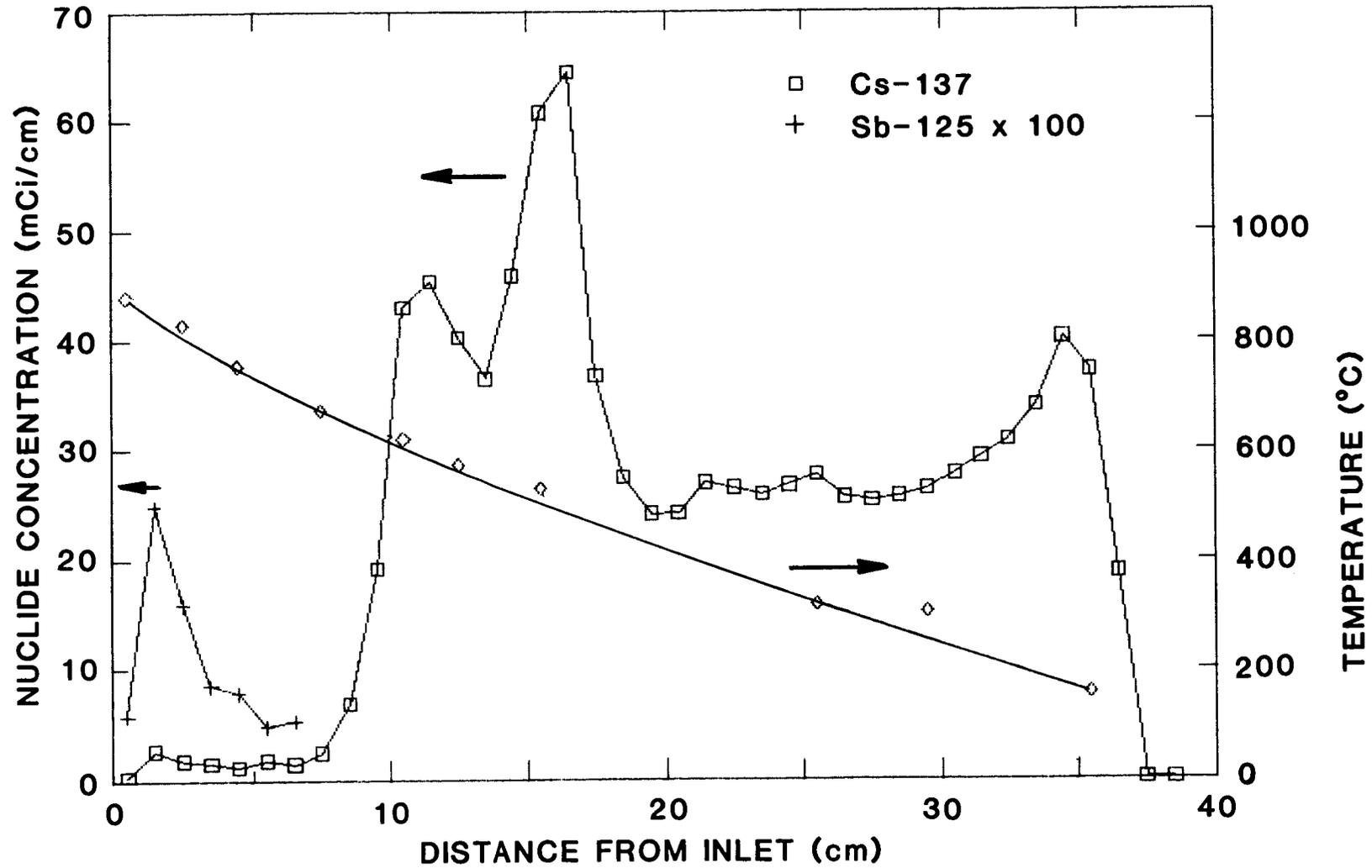


Fig. 12. Distributions of <sup>137</sup>Cs and <sup>125</sup>Sb in TGT A.

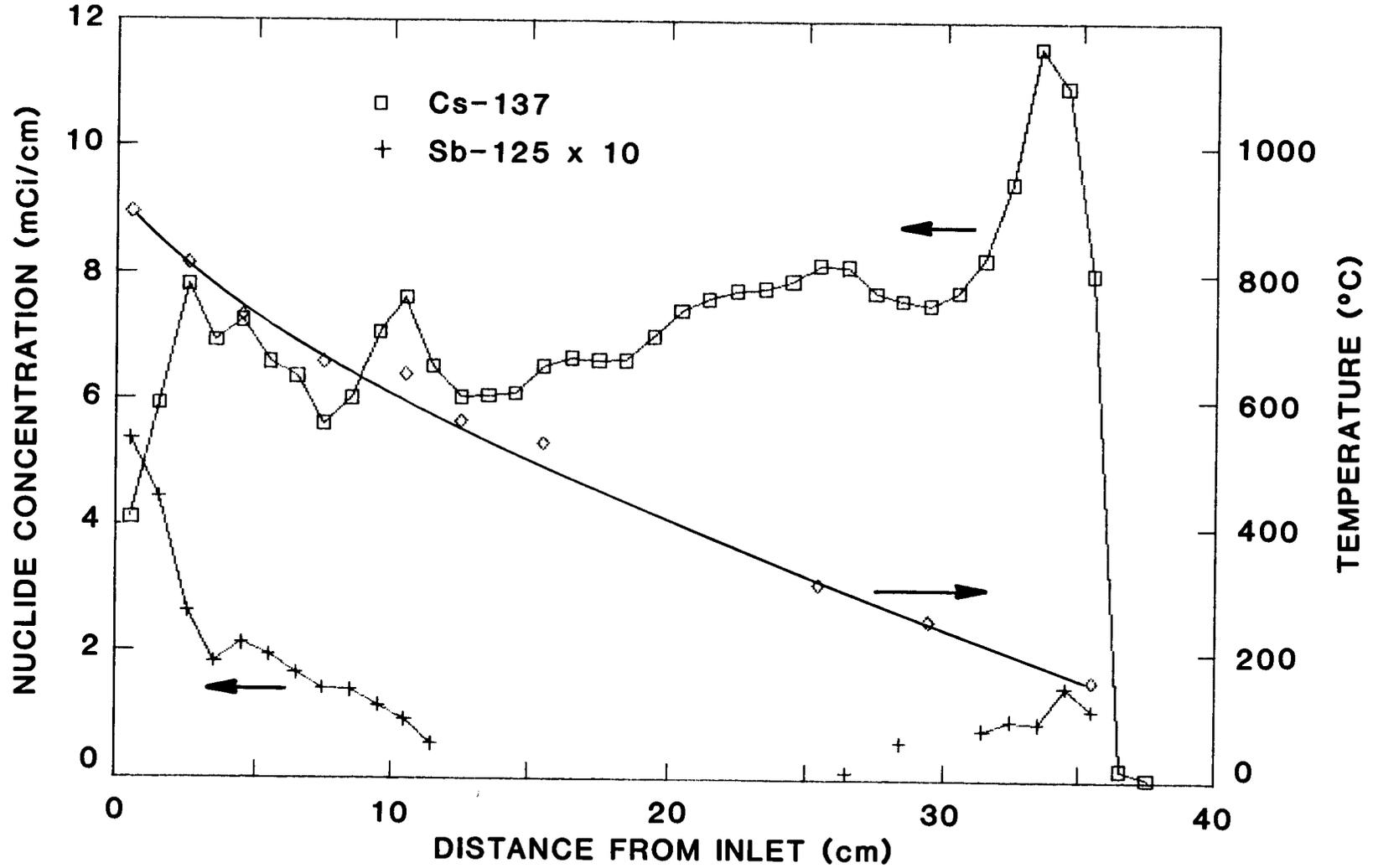


Fig. 13. Distributions of <sup>137</sup>Cs and <sup>125</sup>Sb in TGT B.

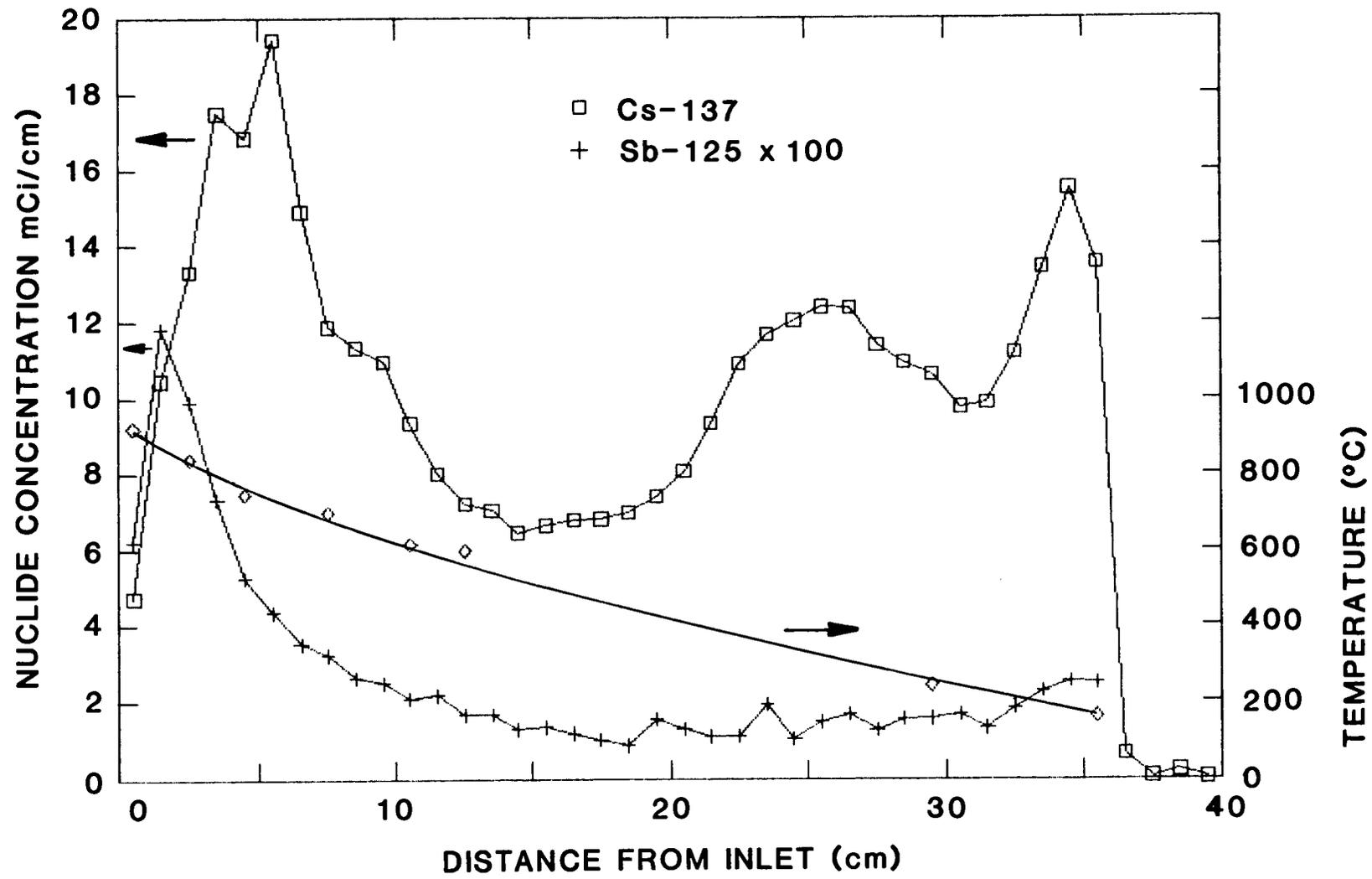


Fig. 14. Distributions of <sup>137</sup>Cs and <sup>125</sup>Sb in TGT C.

very low yield fission product) and very little iodine were found in the SSMS analyses of TGT samples, so there was little opportunity for correlation of these data with the SSMS data (Sect. 4.2.3).

The platinum TGTs were cut into sections of 6 to 15 cm in length and opened to permit visual examination and the collection of smear samples for SSMS analysis. After cutting small samples for SEM examination, the sections were leached with basic solution ( $\text{NH}_4\text{OH}$  and  $\text{H}_2\text{O}_2$ ) to dissolve the iodine forms and hold them in solution, followed with acidic solution ( $\text{HNO}_3$ -HF) to remove other surface deposits. These chemical treatments removed ~99% of the highly radioactive  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , allowing accurate gamma spectrometric analysis of radionuclides that had alloyed with the platinum, presumably from the elemental vapor phase. The distributions of  $^{137}\text{Cs}$ ,  $^{125}\text{Sb}$ , and  $^{110\text{m}}\text{Ag}$  along the three TGTs are shown in Figs. 15 to 17 — the log of the average linear concentration ( $\mu\text{g}/\text{cm}$ ) of each fission product on each TGT section is plotted vs distance from the TGT inlet. Although there are appreciable scatter of the data, it is interesting to note that the results for all three elements show a similar rate of decrease in concentration along the TGT. The diagonal line on each figure indicates a factor of 1000 decrease in concentration over 40 cm, which is somewhat greater than the 36-cm length of the tube. These approximately exponential decreases in concentration indicate rapid deposition, as would occur in condensation from the vapor phase.

Small samples of TGT material were examined by SEM/EDX. These studies provided very interesting views of the morphology of deposits, but very little information about fission product release and behavior.<sup>20</sup> Cesium was generally present in these deposits; some crystals that had been deposited at ~600°C were thought to have been  $\text{CsOH}$ . A significant concentration of tellurium was found on one sample. With the exception of tin (a minor component of the Zircaloy cladding that was observed in most deposits), the only additional deposited material appeared to be impurities from the structural ceramics Si and S. Some examples of the deposits examined by SEM from TGT B are shown in Fig. 18. A large range of features was observed, from spheres to thick whiskers, and the principal elemental components were Sn, S, and Cs. The furnace temperature was raised during this portion of the test, resulting in an increase in the silicon and sulfur release rates from the ceramics. The deposits were not very thick, as Pt was always strongly featured in the X-ray spectrum, but the surface had become extremely porous, apparently the result of some complex interactions. Where the reacted surface had flaked away, the Pt beneath was severely altered but did not contain a measurable amount of impurity. Platinum strands appeared to have erupted from grains and then reacted with S and Si, which formed crystallite side growths on the strands. The spherical objects [Fig. 18(a)] may have been enriched in Cs. A further sample from this region was examined after basic and acidic leaches, but no significant changes in appearance were visible.

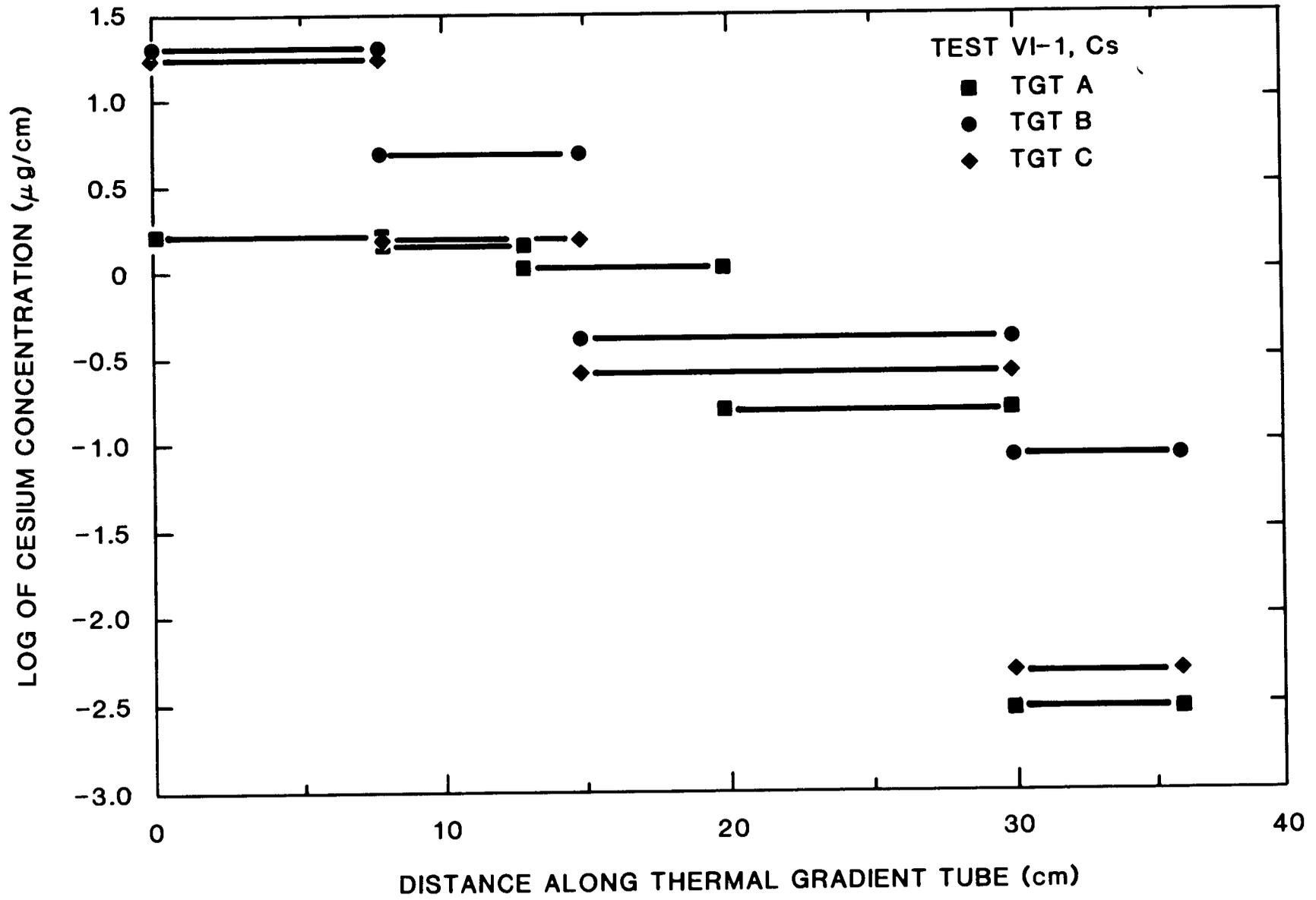


Fig. 15. Concentrations of cesium on TGTs A, B, and C after leaching to remove surface deposits.

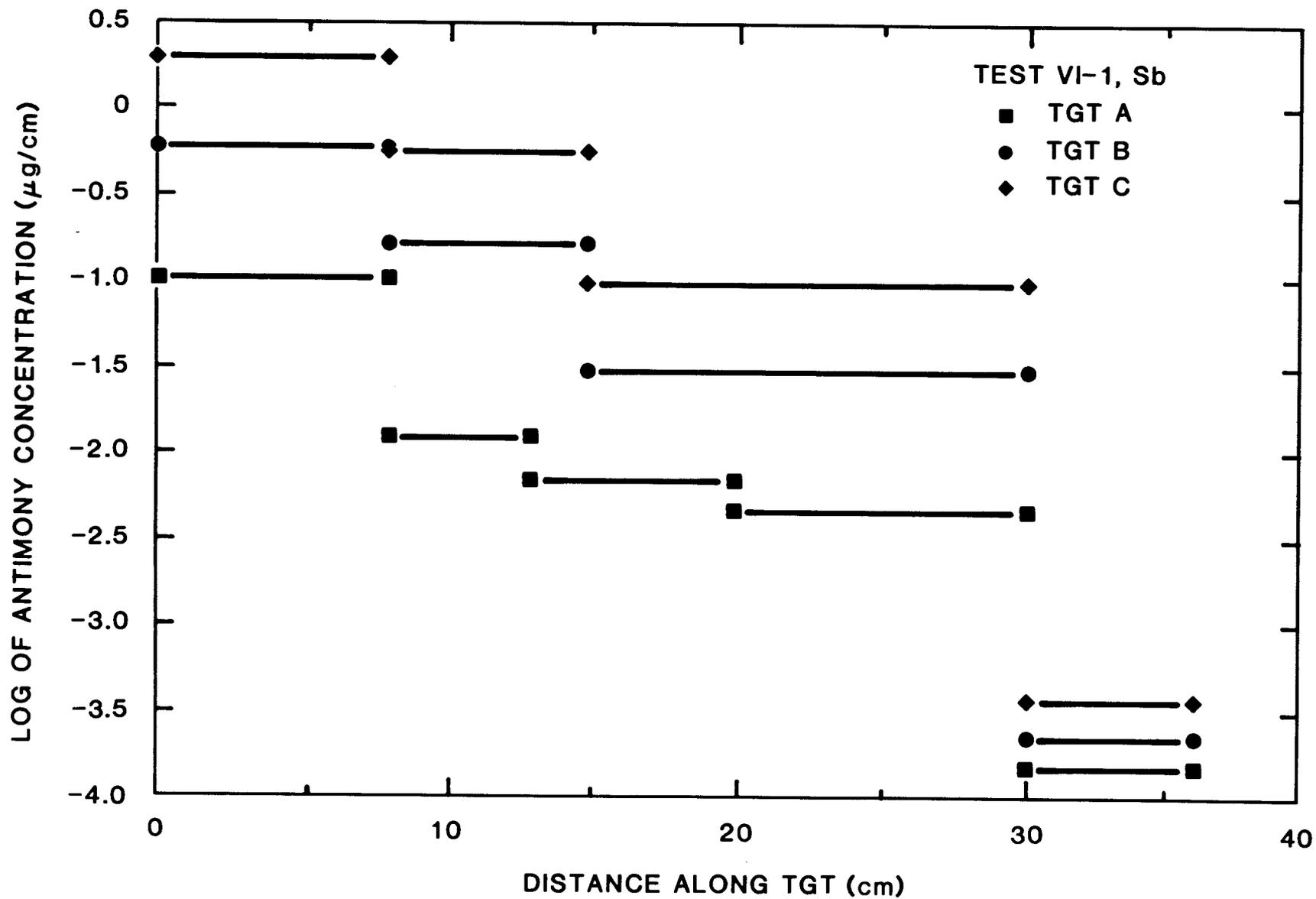


Fig. 16. Concentrations of antimony on TGTs A, B, and C after leaching to remove surface deposits.

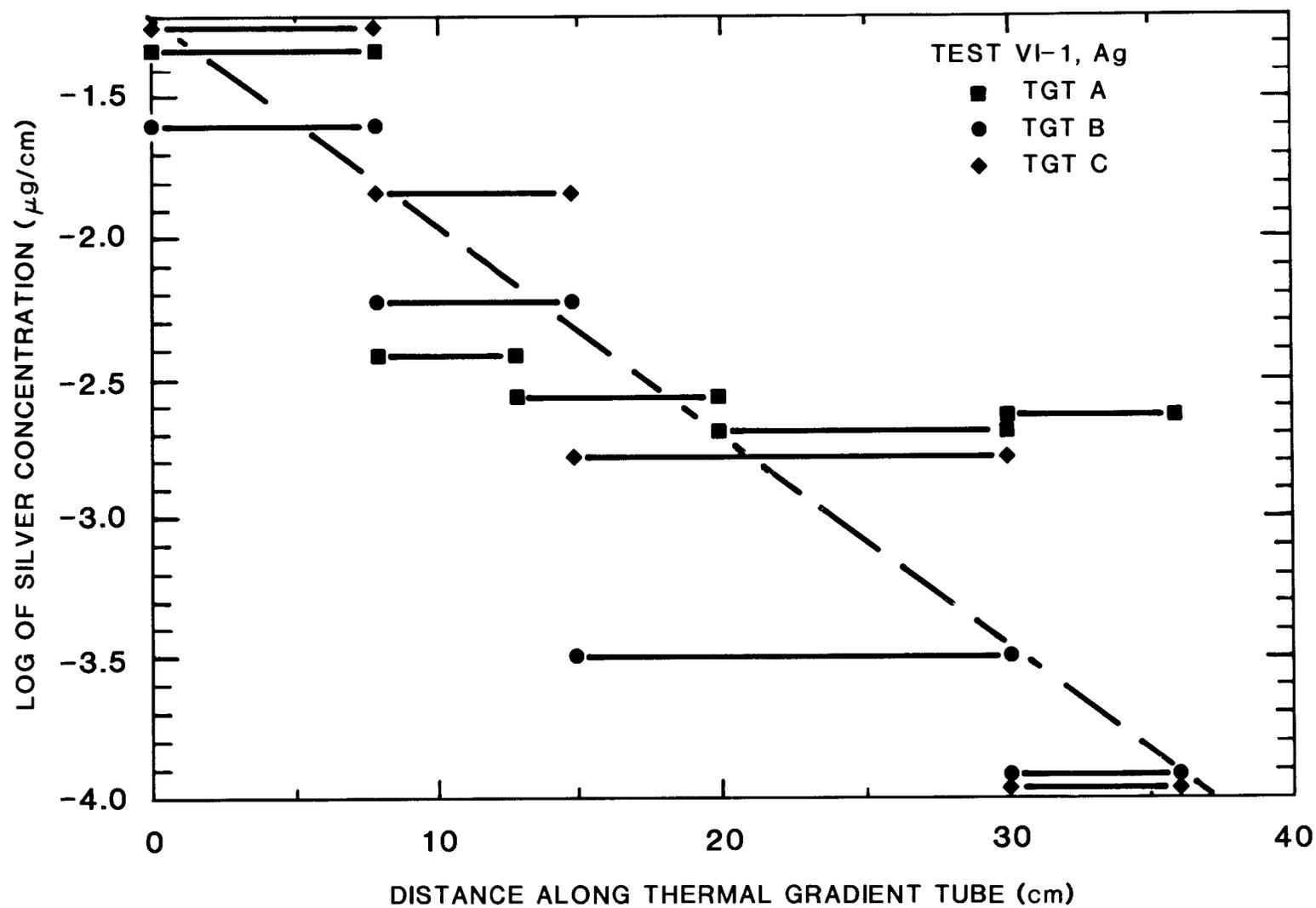


Fig. 17. Concentrations of silver on TGTs A, B, and C after leaching to remove surface deposits.

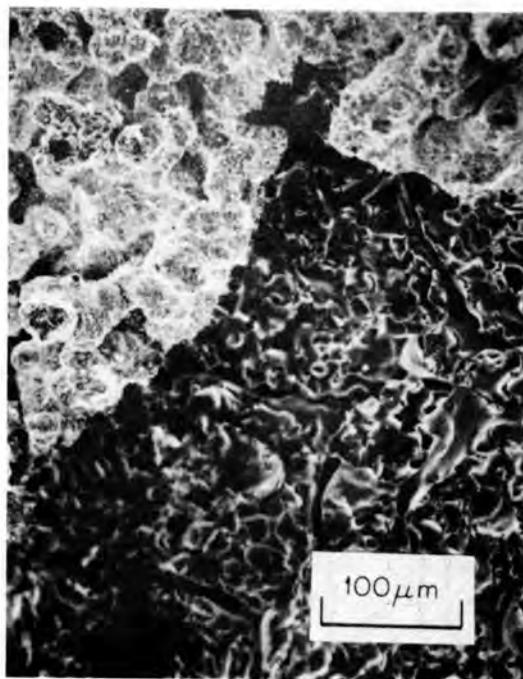
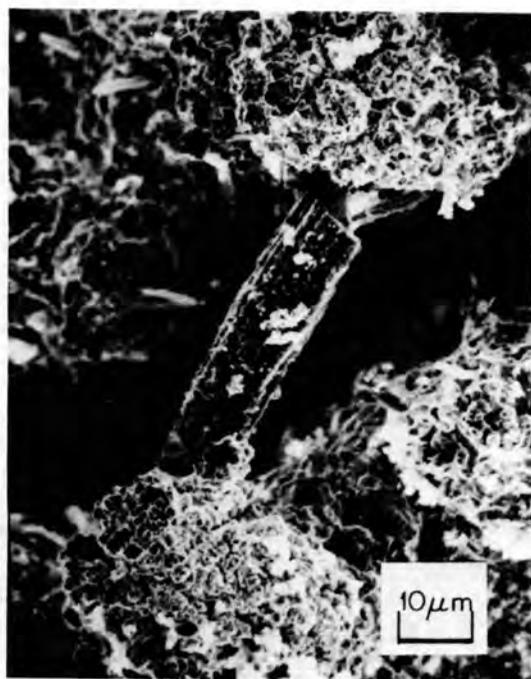
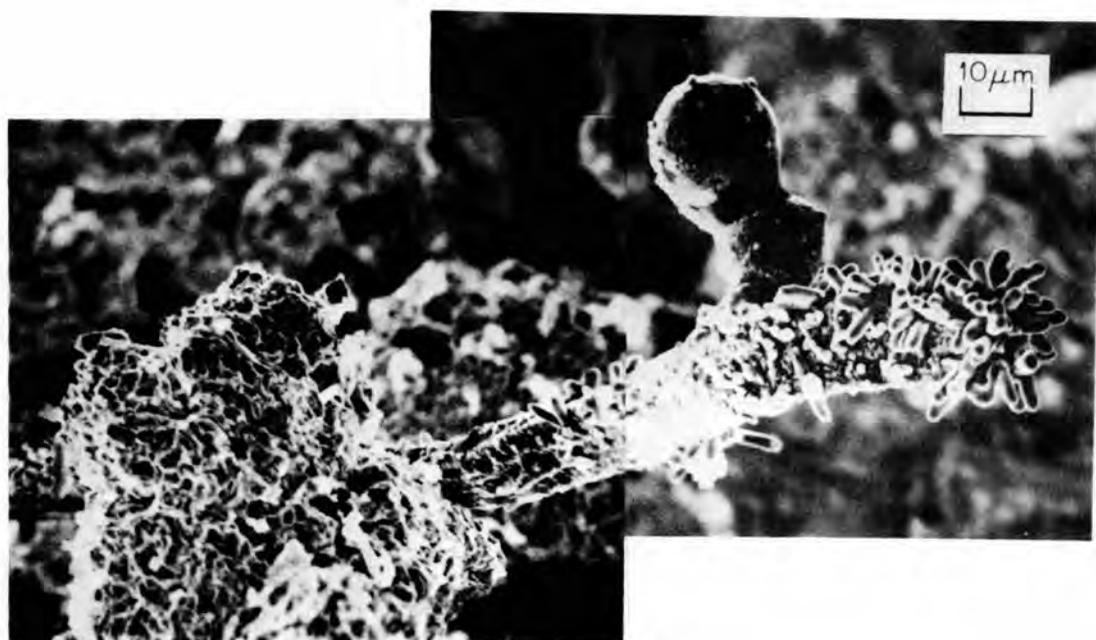


Fig. 18. Photomicrographs of surface deposits at the inlet region of TGT B.

#### 4.2.5 Uranium Release

Since the release and transport of fuel material (mainly U and Pu oxides) provides a potential mechanism for the release of low volatility fission products (in addition to the direct hazard of the heavy metals), various samples were collected and analyzed for uranium. The uranium data, obtained by fluorimetric analysis, are summarized in Table 14. Predictably, very little of the uranium release from the fuel occurred during test phase A at ~2000 K; more than 100 times as much (3.1 mg vs 0.02 mg) occurred during test phase B, which included the heatup from 2020 K to 2300 K and the 5-min period at 2300 K. The total uranium release, however, was only 3.68 mg, ~0.004% of the uranium inventory in the fuel specimen. Larger release fractions would be expected at the high temperatures and longer times are planned for subsequent tests.

Table 14. Uranium release from fuel during test VI-1

Test phase	Fuel conditions		Uranium release	
	Time (min)	Temperature (K)	(mg)	(% of total)
A	20 <sup>a</sup>	2020	0.023	$0.03 \times 10^{-3}$
B	5 <sup>a</sup>	2300	3.14	$3.44 \times 10^{-3}$
C	15 <sup>b</sup>	2300	0.52	$0.57 \times 10^{-3}$
Total			3.68 <sup>c</sup>	$4.04 \times 10^{-3}$

<sup>a</sup>Some additional time to account for heatup period.

<sup>b</sup>Some additional time to account for cooldown.

<sup>c</sup>Because some components were not leached, totals should be considered minimum values.

#### 4.2.6 Masses of Deposits in TGTs and on Filters

Each filter and TGT was weighed before and after the test to determine the mass of material collected during the test. Immediately after disassembly of the filter packages, the filters were inspected and photographed, then packaged for weighing. The filters from Train B are shown in Fig. 19. Both prefilters and the first HEPA filter exhibited deposits, but the last HEPA filter appeared to be unblemished, indicating efficient collection of the aerosols by the previous filters. The masses of material collected at the various locations are listed in Table 15 and illustrated in Fig. 20. As would be expected, the greatest mass was released during the highest temperature period (Train C).



Fig. 19. Appearance of filters from Train B, test VI-1; (a) first prefilter, (b) second prefilter, (c) first HEPA, (d) second HEPA.

Table 15. Vapor and aerosol deposits in test VI-1

	Weight of deposits (g) <sup>a</sup>		
	Train A	Train B	Train C
Thermal gradient tube	0.078	0.101	0.225
Filters:			
Prefilter 1 <sup>b</sup>	0.159	0.182	0.252
Prefilter 2	0.050	0.030	0.029
HEPA 1	0.018	0.007	0.008
HEPA 2	0.018	0.004	0.008
Total filters	0.245	0.223	0.297
Total TGT and filters	0.323	0.324	0.522
Total experiment			1.169
Aerosol concentration (g/m <sup>3</sup> ) <sup>c</sup>	8.1	11.3	9.1

<sup>a</sup>Precision  $\pm 0.003$  g.

<sup>b</sup>Includes estimated mass of deposits in connecting tubes, based on <sup>137</sup>Cs data.

<sup>c</sup>Average value of 423 K, based on total gas flow.

Most of the material deposited in the TGTs is believed to be a result of vapor condensation, and the material deposited on the filters probably was transported primarily as aerosol particles. The average mass concentration of vapor and aerosol during each test phase was calculated from the masses collected and from the total gas flow during that period, as shown in Table 15. These data show that the average mass concentration was highest during Test Phase B, and that this average mass concentration was relatively constant, varying from 8.1 to 11.3 g/m<sup>3</sup> at 423 K.

#### 4.2.7 Fuel Examination

As noted previously, the fuel specimen was cast in epoxy resin to preserve the test geometry, then cut with an abrasive saw for examination of the fuel cross sections. The general appearance of these sections (after only moderate polishing) is shown in Fig. 21. As may be seen, evidence of clad melting is apparent at only one location, ~2 cm from the top end. Otherwise, the cladding appears heavily oxidized with occasional fractures. Two of the above sections were selected for limited metallographic examination. These two sections (nos. 2 and 5 from

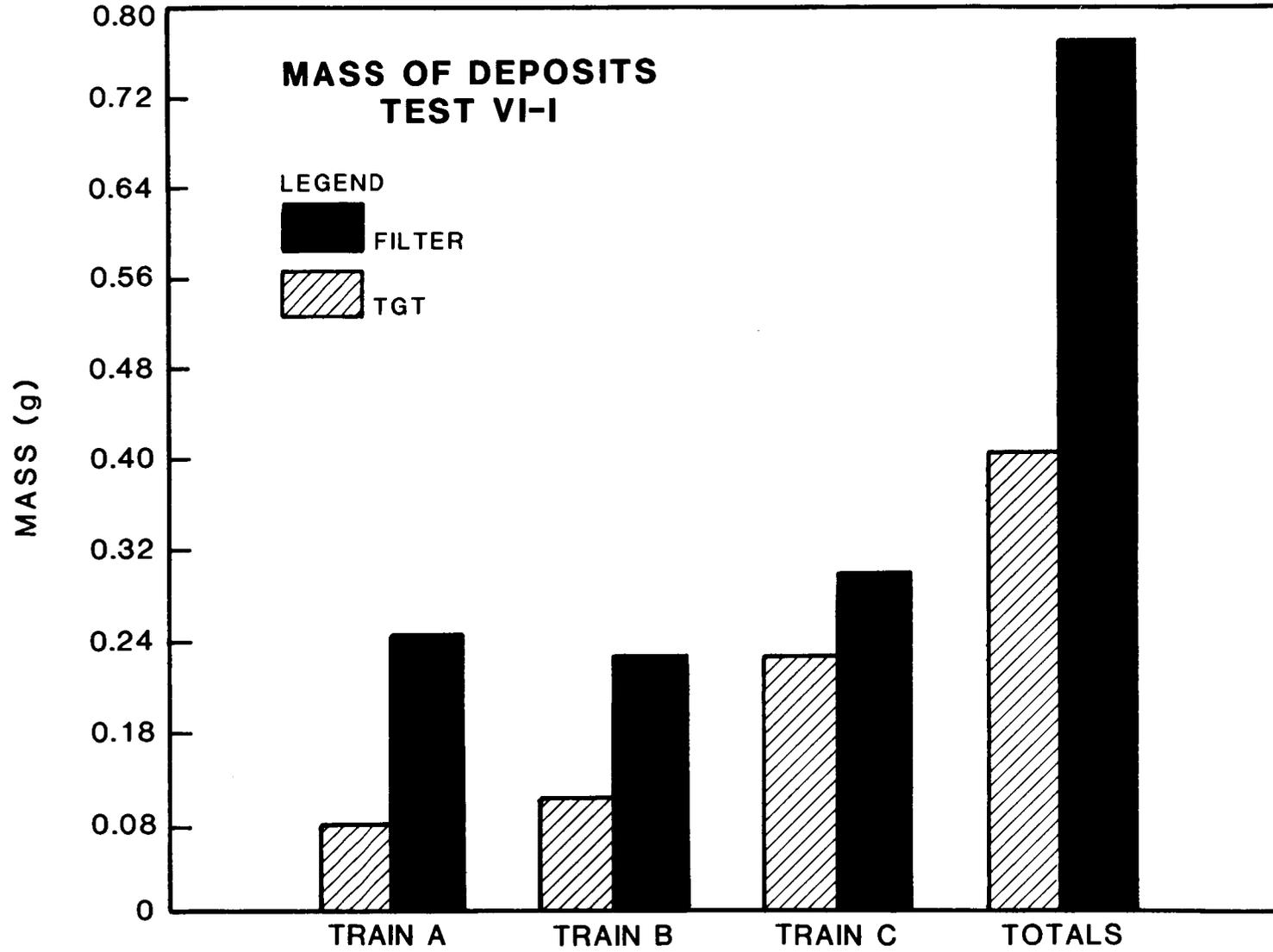


Fig. 20. Distribution of deposits collected in TGTs and on filters in test VI-1.

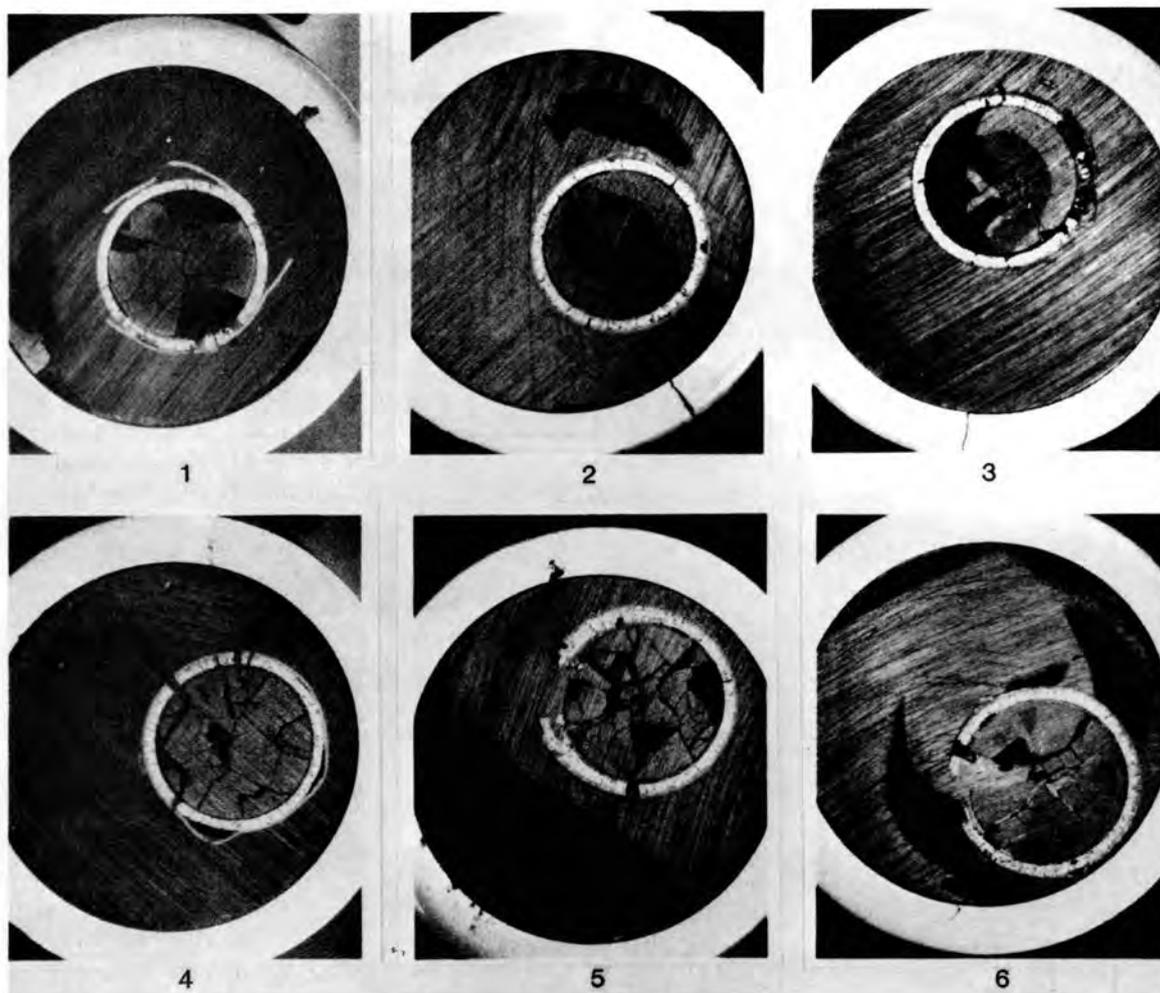


Fig. 21. Cross-sectional views of fuel specimen from test VI-1 at ~2-cm intervals; section 1 was near bottom end, and section 6 was near top end of specimen.

Fig. 21), after thorough polishing, are shown in Fig. 22. The cladding in both sections was completely oxidized and contained large grains (see Fig. 23), as would be expected under the severe conditions of this test. The minimal examination of the  $UO_2$  fuel revealed extensive porosity, a result of the high temperature test, but no unusual features. As shown in Fig. 23, several areas of minor fuel-clad interaction were found, but these did not appear to be severe enough to influence cladding failure.

##### 5. COMPARISON OF RELEASE DATA WITH PREVIOUS RESULTS

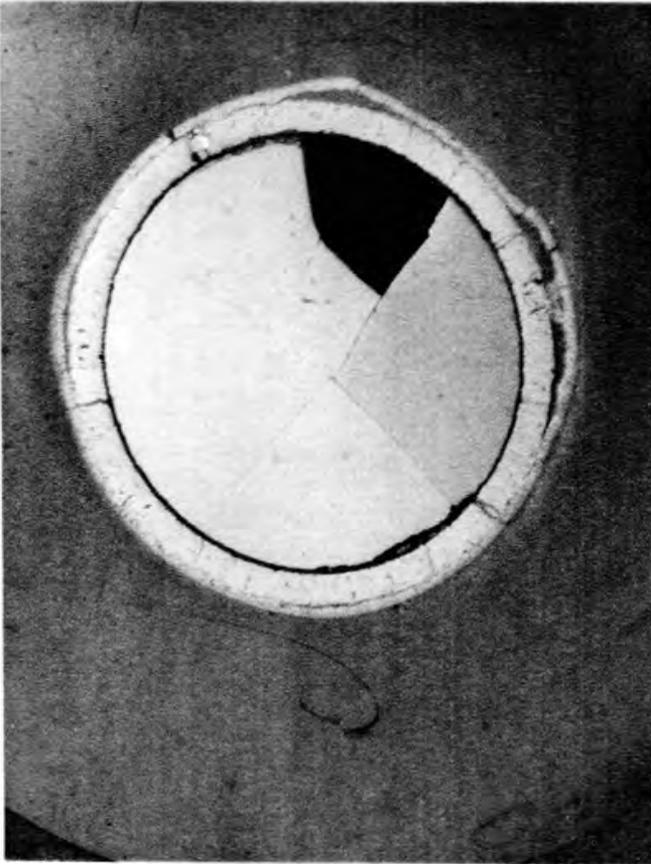
The fission product release data from this test (VI-1) have been compared with the results from earlier related experiments, and also with a comprehensive NRC review of all relevant fission product release data.<sup>21</sup> Since this was the first test in a new vertically oriented furnace, we were particularly interested in comparing these results with the results from similar tests in a horizontal furnace. Two previous tests in the horizontal furnace were conducted with a similar temperature/time regime; the release data from these tests are compared with the VI-1 data in Table 16. As these data indicate, the fractional releases of Kr, I, and Cs varied widely in these three tests. Although the test temperatures and times are similar, other test parameters were significantly different and probably dominate any effects of furnace orientation. Atmospheric conditions probably are the most important. In tests HI-2 and VI-1, high steam flow rates ( $>1$  L/min) were used, resulting in complete oxidation of the Zircaloy cladding and subjecting the  $UO_2$  fuel to an oxidizing (steam) environment. Conversely, a low steam flow rate ( $\sim 0.4$  L/min) in test HI-5 was not sufficient to completely oxidize the cladding, so the  $UO_2$  fuel was in a reducing environment throughout the test. A second difference was the type of fuel used: very low power H. B. Robinson fuel of  $\sim 0.3\%$   $^{85}Kr$  release in-pile in test HI-2 vs much higher power (and somewhat higher burnup) Oconee fuel of  $\sim 4.1\%$  and  $\sim 0.7\%$   $^{85}Kr$  release in tests HI-5 and VI-1, respectively. Examination of specimens of the HI fuel at Argonne National Laboratory showed that the  $UO_2$  in test HI-2 had experienced some oxidation,\* which is thought to be the dominant factor in the higher volatile fission product release ( $\sim 52\%$ ) compared to tests HI-5 and VI-1.

The release rate of cesium in test VI-1 is compared with similar data from the HI tests and with the NRC review curve in Fig. 24. Because test VI-1 was conducted in three phases, it provided three release rate values for three different effective temperatures. (Effective temperatures, slightly different from the measured temperatures, were calculated based on the time spent at less-than-test temperature.) These three data points (Fig. 24) are comparable to, but indicate a somewhat weaker temperature dependence (lower slope), than indicated by the data from the HI tests. The extent of this difference, however, does not appear to be greater than that explainable by the differences in test procedure. Like most of the HI test data, the cesium release rate in test VI-1 falls

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\*R. V. Strain, Argonne National Laboratory, Personal Communication, January 1986.

(a)



(b)

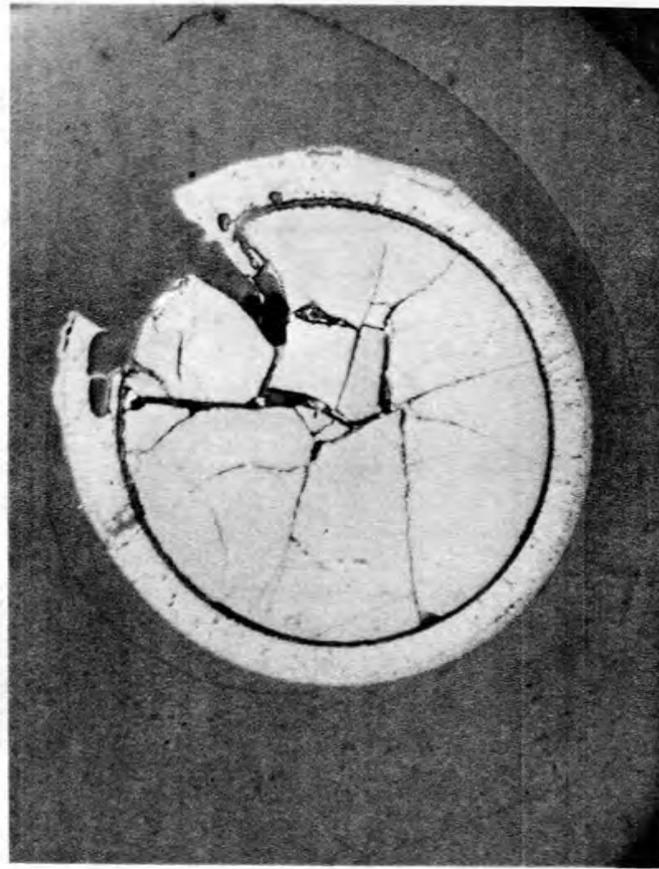


Fig. 22. Appearance of selected fuel sections from VI-1, after polishing for microscopic examination.

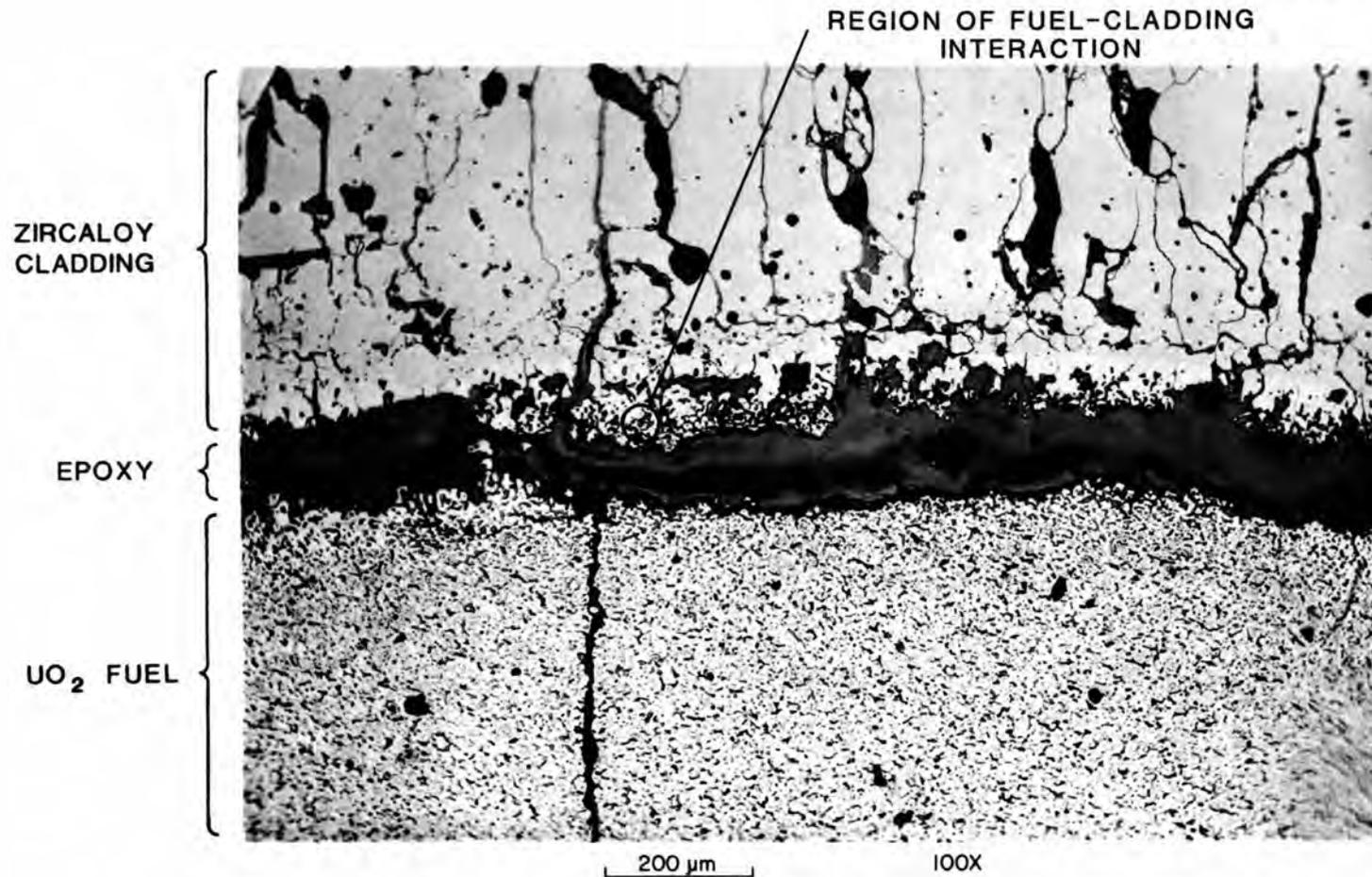


Fig. 23. View of fuel-cladding interface in section 5 of test VI-1 fuel specimen. Note large, columnar grains in oxidized cladding, and extensive fine porosity in UO<sub>2</sub> fuel.

Table 16. Comparison of release data from tests at similar conditions

Test No.	HI-2	HI-5	VI-1A <sup>a</sup>
Temperature (K)	2000	2025	2020
Effective time (min)	22.5	21.5	25
Steam flow rate <sup>b</sup>	High	Low	High
Release (%)			
<sup>85</sup> Kr	51.8	19.9	c
<sup>129</sup> I	53.0	22.4	17.7
<sup>137</sup> Cs	50.5	20.3	28.7

<sup>a</sup>Test Phase A only; Phases B and C were at higher temperatures.

<sup>b</sup>At high steam flow rate (~1 L/min) cladding was completely oxidized; at low steam flow rate (~0.3 L/min) cladding was only partially oxidized.

<sup>c</sup>No intermediate time measurement of <sup>85</sup>Kr in test VI-1, but total release was similar to <sup>137</sup>Cs value.

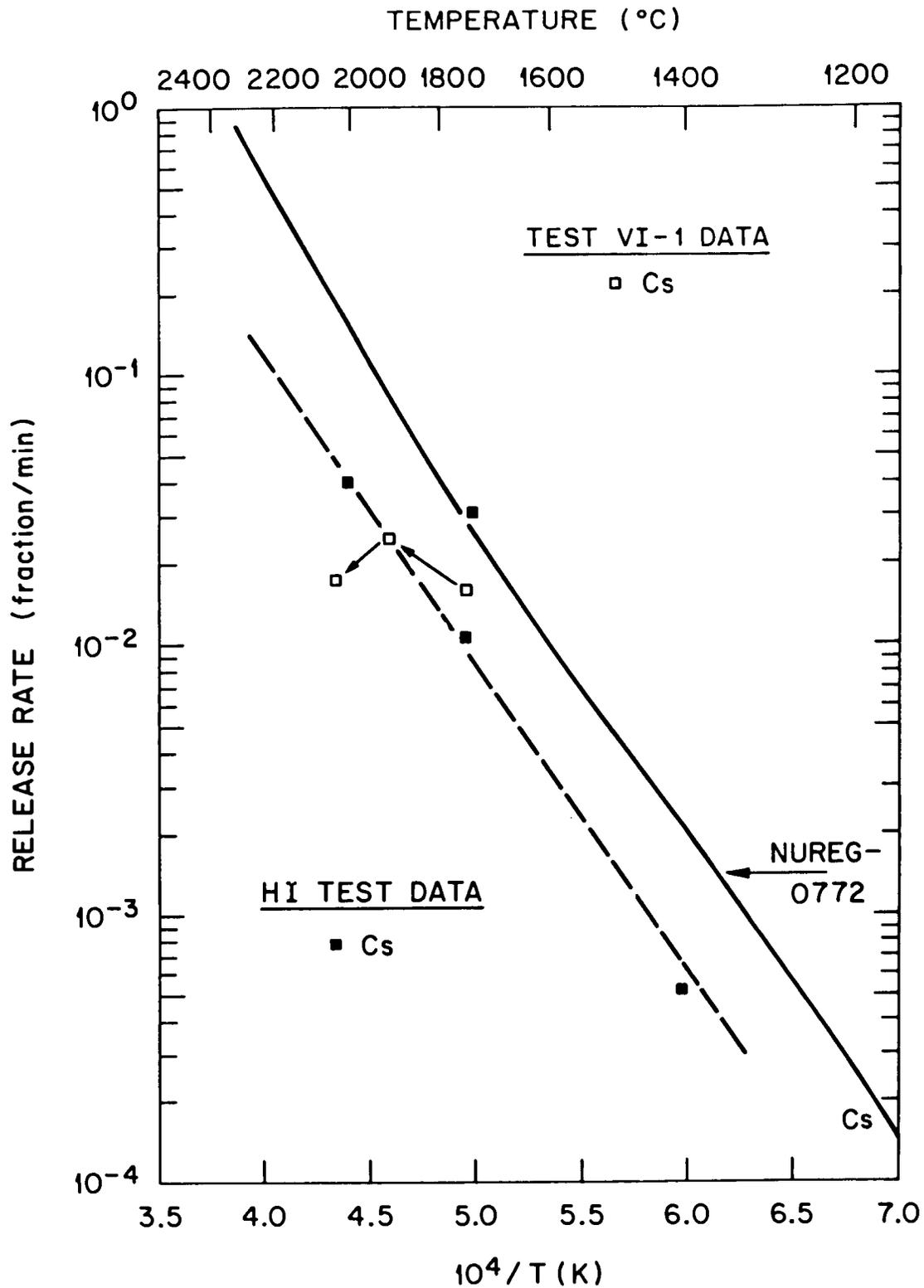


Fig. 24. Comparison of release-rate coefficients from test VI-1, from HI tests, and from NUREG-0772 (ref. 23).

significantly (a factor of ~3) below the NUREG-0772 curve.<sup>21</sup> The release fractions for  $^{110m}\text{Ag}$  and  $^{125}\text{Sb}$  (33.2 and 28.0% respectively in Table 9) are higher than those that have been observed in any previous tests. Both of these nuclides are of relatively low yield, however, and their nuclear decay characteristics result in lower analysis precision than for  $^{85}\text{Kr}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$ . Based on the sparse available data, we suspect that the release of these elements (Ag and Sb) is sensitive to the extent of cladding oxidation, as has been observed in the case of tellurium. Methods for the determination of tellurium release by neutron activation analysis are being developed, but have not yet produced reliable data for this test.

## 6. CONCLUSIONS

As noted previously, a thorough interpretation of the results of test VI-1 are beyond the scope of this report. However, several significant observations are appropriate.

1. The new vertical furnace operated well and the planned test conditions were accomplished. All components of the expanded fission product collection system provided useful data. The apparatus for the continuous measurement of hydrogen operated very well, proving its usefulness as an aid in the interpretation of test results. Consequently, the test and the pretest apparatus modifications are considered successful.
2. Techniques for determining the inventories of fission products in the irradiated fuel were significantly improved, thereby allowing more accurate release measurements. Detailed gamma spectrometry of the specimen, accompanied by improved shielding corrections, provided good data for the long-lived gamma emitters such as  $^{106}\text{Eu}$ ,  $^{125}\text{Sb}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{144}\text{Ce}$ , and  $^{154}\text{Eu}$ . Fitting of neodymium and uranium isotopic data (from mass spectroscopic analysis) and the  $^{137}\text{Cs}$  data with results from ORIGEN2 calculations produced improved values for other important fission product nuclides, especially for  $^{85}\text{Kr}$ ,  $^{110m}\text{Ag}$ , and  $^{129}\text{I}$ , and for all fission product elements.
3. The total release values for the more volatile fission products — Kr, Ag, Sb, I, and Cs — were determined for the specific conditions of this test. Comparison of these data with the results from similar previous tests shows significant differences but experimental differences appear to explain the most pronounced discrepancies. Higher release values for Ag and Sb (compared to previous tests) indicate an area deserving of further investigation.
4. The oxidation behavior of the Zircaloy cladding, as indicated by continuous measurement of hydrogen generation, was in good agreement with a previously developed model, indicating an adequate understanding of this reaction that significantly influences the behavior of some fission products.

5. Posttest examination of the fuel specimen showed that the cladding was completely oxidized and that minimal fuel-cladding interactions had occurred, verifying that the planned test conditions had been achieved.

## 7. REFERENCES

1. R. A. Lorenz, J. L. Collins, A. P. Malinauskas, O. L. Kirkland, and R. L. Towns, Fission Product Release from Highly Irradiated LWR Fuel, NUREG/CR-0722 (ORNL/NUREG/TM-287/R2), Oak Ridge National Laboratory, February 1980.
2. R. A. Lorenz, J. L. Collins, A. P. Malinauskas, M. F. Osborne and R. L. Towns, Fission Product Release from Highly Irradiated LWR Fuel Heated to 1300-1600°C in Steam, NUREG/CR-1386 (ORNL/NUREG/TM-346), Oak Ridge National Laboratory, November 1980.
3. R. A. Lorenz, J. L. Collins, M. F. Osborne, R. L. Towns, and A. P. Malinauskas, Fission Product Release from BWR Fuel Under LOCA Conditions, NUREG/CR-1773 (ORNL/NUREG/TM-388), Oak Ridge National Laboratory, July 1981.
4. R. A. Lorenz, J. L. Collins and A. P. Malinauskas, "Fission Product Source Terms for the Light Water Reactor Loss-of-Coolant Accident," Nucl. Technol. 46, 404-10 (1979).
5. M. F. Osborne, J. L. Collins, P. A. Haas, R. A. Lorenz, J. R. Travis, and C. S. Webster, Design and Final Safety Analysis Report for Vertical Furnace Fission Product Release Apparatus in Hot Cell B, Building 4501, NUREG/CR-4332 (ORNL/TM-9720), Oak Ridge National Laboratory, March 1986.
6. M. F. Osborne, J. L. Collins, R. A. Lorenz, J. R. Travis, and C. S. Webster, Design, Construction, and Testing of a 2000°C Furnace and Fission Product Collection System, NUREG/CR-3715 (ORNL/TM-9135), Oak Ridge National Laboratory, September 1984.
7. M. F. Osborne, R. A. Lorenz, J. R. Travis and C. S. Webster, Data Summary Report for Fission Product Release Test HI-1, NUREG/CR-2928 (ORNL/TM-8500), Oak Ridge National Laboratory, December 1982.
8. M. F. Osborne, R. A. Lorenz, J. R. Travis, C. S. Webster, and K. S. Norwood, Data Summary Report for Fission Product Release Test HI-2, NUREG/CR-3171 (ORNL/TM-8667), Oak Ridge National Laboratory, February 1984.
9. M. F. Osborne, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-3, NUREG/CR-3335 (ORNL/TM-8793), Oak Ridge National Laboratory, April 1984.

10. M. F. Osborne, J. L. Collins, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-4, NUREG/CR-3600 (ORNL/TM-9011), Oak Ridge National Laboratory, June 1984.
11. M. F. Osborne, J. L. Collins, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-5, NUREG/CR-4037 (ORNL/TM-9437), Oak Ridge National Laboratory, May 1985.
12. M. F. Osborne, J. L. Collins, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-6, NUREG/CR-4043 (ORNL/TM-9443), Oak Ridge National Laboratory, September 1985.
13. G. M. Bain and W. A. McInteer, Data Package for High Burnup Fuel Samples, LRC 4063, Lynchburg Research Center, Babcock and Wilcox Co., January 1983.
14. A. G. Croff, ORIGEN2 - A Revised and Updated Version of the Oak Ridge Isotope Generation and Depletion Code, ORNL/5621, Oak Ridge National Laboratory, July 1980.
15. Toshiyuki Yamashita, Steam Oxidation of Zircaloy Cladding in the ORNL Fission Product Release Tests, NUREG/CR-4777 (ORNL/TM-10272), Oak Ridge National Laboratory, March 1988.
16. L. Baker and L. C. Just, ANL-6548, Argonne National Laboratory, 1962.
17. V. F. Urbanic and T. R. Heidrick, J. Nucl. Mater. 75, 251 (1978).
18. J. L. Collins, M. F. Osborne, and R. A. Lorenz, "Fission Product Tellurium Release Behavior Under Severe Light Water Reactor Accident Conditions," Nucl. Technol. 77(1), 18-31 (April 1987).
19. K. S. Norwood, An Assessment of Thermal Gradient Tube Results from the HI Series of Fission Product Release Tests, NUREG/CR-4105 (ORNL/TM-9506), Oak Ridge National Laboratory, March 1985.
20. S. K. Wisbey, Preliminary Studies of the Morphology of Thermal Gradient Tube Deposits for Fission Product Release Experiments, NUREG/CR-4778 (ORNL/TM-10273), Oak Ridge National Laboratory, March 1988.
21. U. S. Nuclear Regulatory Commission, Technical Bases for Estimating Fission Product Behavior During LWR Accidents, NUREG-0772, June 1981.

## APPENDIX. GENERAL PROCEDURE FOR FISSION PRODUCT RELEASE TESTS

A. Days before test

1. Prepare system — furnace ceramics, TGTs, filter packages, tools, traps, etc., in both Cell B and Cell D.
2. Check operation of heaters, thermocouples, recorders, flowmeters, etc. Measure pressure drop between susceptor and fuel regions.
3. Prepare fuel specimen in Cell D for transfer into Cell B. Do gamma spectrometry in Room 220: whole rod through lead and unshielded 1-cm scan.
4. Verify in-cell remote handling procedures and furnace loading/unloading.
5. Collect fuel specimen data and specify test operating conditions, planned temperature cycles, etc.
6. Fill steam generator.
7. Assemble, evacuate, and leak check the test apparatus.
8. Check operation of all radiation detectors; calibrate with standard or accurately known sources.
9. Prepare and check posttest examination equipment in Hot Cells A, B, and D.
10. Verify sealing of furnace and preheat to purify ceramics.

B. Day of test

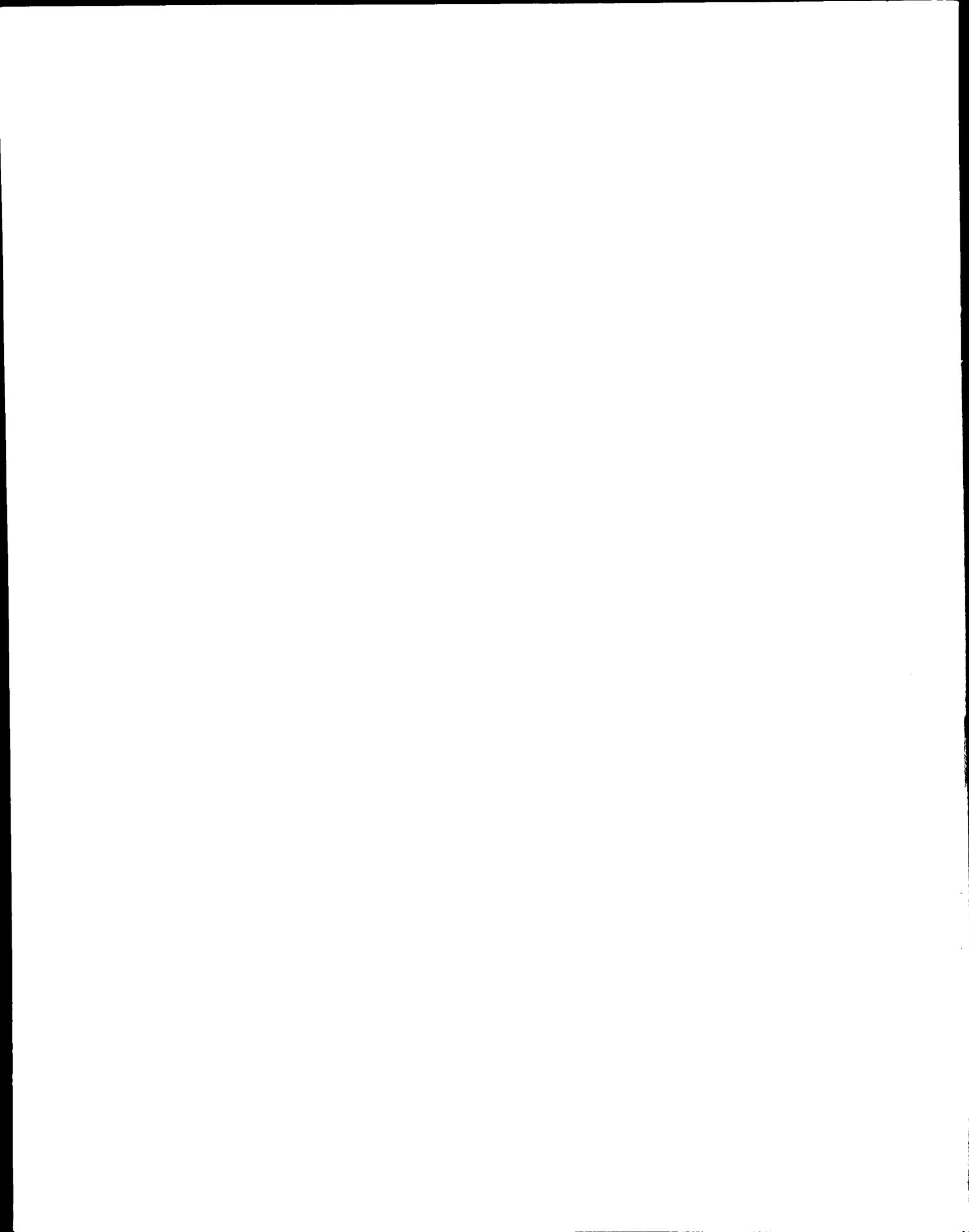
1. Recheck all radiation detectors, multichannel analyzer, data recording program, and data acquisition system.
2. Close and lock all cells; tape doors and set Cell B vacuum (-1.0 in. H<sub>2</sub>O).
3. Bring in fuel specimen, install in furnace, and connect lines.
4. Evacuate, purge, and leak check the test apparatus with steam line heat on.
5. Close box, set vacuum to -10 in. H<sub>2</sub>O and 40 L/min airflow.
6. Set helium flow rates at planned values.
7. Preheat furnace slowly to ~150°C.

8. Heat steam generator, begin flow to vent at 0.3 L/min helium (MFM-3) and  $T_{SG} = 95.6^{\circ}\text{C}$  (for 1.0 L/min).
9. Verify proper TGT temperatures.
10. Begin steam flow through furnace; record flow rates and back pressures, and adjust TGT furnaces as needed.
11. Set data acquisition system (DAS) to 1-min recording cycle.
12. When all flows stabilize (minimum 5-min steam flow), begin planned heatup cycle,  $\sim 250^{\circ}\text{C}$  to test temperature.
13. Considering correction factors for thermocouple and optical pyrometer, reduce power when test temperature is reached to maintain for desired time period.
14. Monitor all instruments and chart recorders and mark at regular intervals during test, each experimentalist having specific responsibilities.
15. At end of planned period, switch collection trains and heat to new temperature, or reduce as planned.
16. Turn power down or off at end of test period, continuing steam flow until temperature drops below  $1200^{\circ}\text{C}$ .
17. Turn steam generator power off and switch steam flow to vent, with MFM-3 helium bypassing steam generator.
18. Continue helium flow through furnace  $\sim 30$  min to dry system, then turn off power to line heaters and TGT and filter furnaces.
19. Reduce helium flow to MFM-2 = 0 and MFM-3 = 0.1 L/min, and bypass condenser, dryer, and cold charcoal traps.
20. Turn on vacuum pump and reduce pressures in cold traps to  $\sim 0.5$  atm; then close individually and remove from system for analysis.

C. Days following test

1. Disconnect filter packages, count individually with NaI detector, and transfer from Cell B to Cell D for disassembly.
2. Record another set of gamma spectra for background correction.
3. Reduce vacuum on containment box, open, and inspect furnace assembly; begin off-gas flow to prefilter work table and position at top end of furnace.

4. Remove TGT-to-filter lines.
5. Remove top flange from furnace, including contained TGT liners, load into transfer/storage can, and transfer to Cell D.
6. Inspect top end of furnace, viewing fuel specimen to the extent possible; photograph if feasible.
7. Pour epoxy to encase fuel in a series of at least three pours, observing after each.
8. Remove fuel assembly, can, and transfer to Cell D for further examination and gamma analysis.
9. Do preliminary remote cleanup of Cell B, transfer all components to be analyzed to Cell A or D.
10. Complete gamma scanning and analysis of fuel, TGTs, filters, etc.
11. Proceed with planned disassembly, sampling, leaching, and analysis of all components.



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5 AUTHOR(S)  M. F. Osborne, J. L. Collins, R. A. Lorenz, J. R. Travis, C. S. Webster, and T. Yamashita*		6 DATE REPORT ISSUED		MONTH June	YEAR 1989
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				13 ABSTRACT (200 words or less)  The first in a series of high-temperature fission product release tests in a new vertical test apparatus was conducted in flowing steam. The test specimen was a 15.2-cm-long section of a fuel rod from the Oconee 1 PWR; it had been irradiated to a burnup of ~42 MWd/kg. Using an induction furnace, it was heated under simulated LWR accident conditions - 20 min at 2000 K and 20 min at 2300 K - in a hot cell-mounted test apparatus. Posttest inspection showed severe oxidation but only minimal fragmentation of the fuel specimen; cladding melting was apparent only near the top end. Based on fission product inventories measured in the fuel and/or calculated by ORIGEN, analyses of test components showed total releases from the fuel of 47% for <sup>85</sup> Kr, 33% for <sup>125</sup> Sb, 37% for <sup>129</sup> I, 84% for <sup>110m</sup> Ag, and 63% for <sup>137</sup> Cs. Large fractions (36% and 30%, respectively) of the released <sup>110m</sup> Ag and <sup>125</sup> Sb were retained in the furnace above the fuel. Pretest and posttest analysis of the fuel specimen indicated a <sup>134</sup> Cs release of 65%, which is very good agreement with the <sup>137</sup> Cs value.  The first phase (2000 K) of this test was conducted at the same conditions as test HI-2. However, the releases of Cs, I, and Kr were much less than those in HI-2. Variations in fuel characteristics (H. B. Robinson in test HI-2 vs Oconee in test VI-1) are believed to be major factors in this difference, and the large axial crack that occurred in HI-2 probably was a significant factor.	
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